

Ottawa Hull K1A 0C9

(21) (A1) 2,158,996 (86) 1994/03/14 (43) 1994/10/13

- 6
  (51) Int.Cl. C07D 471/04; C07D 487/04; C07F 9/547; A01N 43/90; A01N 57/24
- (19) (CA) APPLICATION FOR CANADIAN PATENT (12)
- (54) Substituted Heteroannulated Imidazoles and Their Use as Herbicides
- (72) Bassner, Bernd Germany (Federal Republic of);
  Lieb, Folker Germany (Federal Republic of);
  Santel, Hans-Joachim U.S.A.;
  Dollinger, Markus Germany (Federal Republic of);
- (71) Bayer Aktiengesellschaft Germany (Federal Republic of)
  ;
- (30) (DE) P 43 09 969.6 1993/03/26
- (57) 10 Claims

Notice: This application is as filed and may therefore contain an incomplete specification.





#### (57) Abstract

New substituted heteroannulated imidazoles having general formula (I), in which R<sup>1</sup>, R<sup>2</sup>, R<sup>3</sup>, A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> and A<sup>4</sup> have the meanings given in the description, are disclosed, as well as a process for preparing the same and their use as herbicides.

2158996

BAYER AKTIENGESELLSCHAFT

51368 Leverkusen

Konzemzentrale RP

Patente Konzem

WA/HB

## FILE, PIN IN THIS ASSENDED TEXT-TRANSLATION

# SUBSTITUTED HETERO-FUSED IMIDAZOLES AND THEIR USE AS HERBICIDES

The invention relates to new substituted hetero-fused imidazoles, to a process for their preparation, and to their use as herbicides.

It is known that certain benzimidazoles have insecticidal properties, but nothing has been disclosed about the use of hetero-fused imidazoles as herbicides.

There have now been found new substituted hetero-fused imidazoles of the general formula (I)

$$\begin{array}{c|c}
A & A \\
\downarrow_{2} & N \\
N & R^{3}
\end{array}$$

$$\begin{array}{c|c}
CH-R^{1} \\
\downarrow_{2} \\
R
\end{array}$$
(1)

in which

15

- R<sup>1</sup> represents hydrogen or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl, alkoxy and aryl,
- R<sup>2</sup> represents hydroxyl, cyano or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of

Le A 29 565

### 2158996

alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, amino, aminocarbonyl, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy, dialkoxyphosphonyl, (hetero)aryl, (hetero)arylcarbonyl, (hetero)arylcarbonyloxy and (hetero)arylaminocarbonylamino-carbonyloxy,

- 5 R³ represents cyano, halogen or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl, alkenyl, alkinyl, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy, alkenyloxy, alkinyloxy, amino, aminocarbonyl and aryl,
- A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> and A<sup>4</sup> in each case represent N(nitrogen), N-CHR<sup>1</sup>R<sup>2</sup> or CX, the heterofused ring having at least one, but not more than two, nitrogen atoms simultaneously and all positional isomers being possible, so that
  - CX1, CX2, CX3 exist in the case of one nitrogen atom and
  - CX<sup>1</sup> and CX<sup>2</sup> exist in the case of two nitrogen atoms, and, when either A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> or A<sup>4</sup> represent N-CHR<sup>1</sup>R<sup>2</sup>, the imidazole ring exists only in monosubstituted form (R<sup>3</sup>),

where

15

20

25

X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> in each case independently of one another represent hydrogen, halogen, cyano, nitro or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl, alkoxy, alkylthio, alkylsulphinyl, alkylsulphonyl and cycloalkyl, or represents hydroxycarbonyl, alkylcarbonyl, alkoxycarbonyl, cycloalkyloxycarbonyl, or represents in each case optionally substituted amino or aminocarbonyl, or represents in each case optionally substituted aryl, aryloxy, arylthio, arylsulphinyl, arylsulphonyl, arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylazo or arylthiomethylsulphonyl, but where at least one of the substitutents X<sup>1</sup>, X<sup>2</sup> or X<sup>3</sup> represents halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl, halogenoalkylsulphonyl, or represents

optionally substituted fused dioxyalkylene, or represents hydroxycarbonyl, alkylcarbonyl, alkoxycarbonyl, cycloalkyloxycarbonyl, or represents in each case optionally substituted amino or aminocarbonyl, or represents in each case optionally substituted aryl, arylthio, arylsulphinyl, arylsulphonyl, arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylazo or arylthiomethylsulphonyl.

Depending on the nature and number of substitutents, the compounds of the formula (I) can, if appropriate, exist as geometric and/or optical isomers or regioisomers or variously composed isomer mixtures of these, but also in the form of positional isomers, for example in the following variations:

or, for example, the following variations are also possible:

5

$$X^{2} \xrightarrow{X^{1}} N \xrightarrow{R^{2}} R^{3} \xrightarrow{R^{1}R^{2}} K \xrightarrow{X^{2}} N \xrightarrow{R^{1}R^{2}} K \xrightarrow{X^{2}} N \xrightarrow{R^{1}R^{2}} K \xrightarrow{X^{2}} N \xrightarrow{X^{2}} K \xrightarrow{X^{2$$

Furthermore, it has been found that the new substituted hetero-fused imidazoles of the general formula (I)

$$\begin{array}{c|c}
A^{3} & A^{4} \\
\downarrow_{2} & N \\
N & CH-R^{1} \\
\downarrow_{2} & R
\end{array}$$
(I)

in which

5

10

15

- R<sup>1</sup> represents hydrogen or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl, alkoxy and aryl,
- R<sup>2</sup> represents hydroxyl, cyano or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, amino, aminocarbonyl, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy, dialkoxyphosphonyl, (hetero)aryl, (hetero)arylcarbonyl, (hetero)arylcarbonyloxy, (hetero)arylcarbonyloxy, and (hetero)arylaminocarbonylaminocarbonyloxy,
  - R<sup>3</sup> represents cyano, halogen or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl, alkenyl, alkinyl, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy, alkenyloxy, alkinyloxy, amino, aminocarbonyl and aryl,

A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> and A<sup>4</sup> in each case represent N(nitrogen), N-CHR<sup>1</sup>R<sup>2</sup> or CX, the

Le A 29 565

- 4 -

heterofused ring having at least one, but not more than two, nitrogen atoms simultaneously and all positional isomers being possible, so that

CX1, CX2, CX3 exist in the case of one nitrogen atom and

CX<sup>1</sup> and CX<sup>2</sup> exist in the case of two nitrogen atoms, and, when either A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> or

A<sup>4</sup> represent N-CHR<sup>1</sup>R<sup>2</sup>, the imidazole ring exists only in monosubstituted form
(R<sup>3</sup>),

where,

X1, X2 and X3 in each case independently of one another represent hydrogen, halogen, cyano, nitro or a straight-chain or branched, in each case optionally 10 unsubstituted or substituted, radical from the series consisting of alkyl, alkoxy, alkylthio, alkylsulphinyl, alkylsulphonyl and cycloalkyl, or represents hydroxycarbonyl, alkylcarbonyl, alkoxycarbonyl, cycloalkyloxycarbonyl, or represents in each case optionally substituted amino or aminocarbonyl, or represents in each case optionally substituted aryl, aryloxy, arylthio, 15 arylsulphinyl, arylsulphonyl, arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylazo or arylthiomethylsulphonyl, but where at least one of the substituents X1, X2 or X3 represents halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl, halogenoalkylsulphonyl, alkylsulphonyl, or represents optionally substituted fused dioxyalkylene, or represents hydroxycarbonyl, 20 alkylcarbonyl, alkoxycarbonyl, cycloalkyloxycarbonyl, or represents in each case optionally substituted amino or aminocarbonyl, or represents in each case optionally substituted aryl, arylthio, arylsulphinyl, arylsulphonyl. arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylazo arylthiomethylsulphonyl,

#### 25 are obtained when

a) 1H-substituted hetero-fused imidazoles of the formula (II)

in which

A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup>, A<sup>4</sup> and R<sup>3</sup> have the abovementioned meanings

are reacted with compounds of the formula (III)

in which

5 M represents a suitable leaving group and

R1 and R2 have the abovementioned meanings,

if appropriate in the presence of a diluent and if appropriate in the presence of a reaction auxiliary.

Finally, it has been found that the new substituted hetero-fused imidazoles of the general formula (I) have good herbicidal activity.

Surprisingly, the new substituted hetero-fused imidazoles of the general formula (I) according to the invention show a considerable herbicidal activity against problem weeds combined with a similarly good tolerance by important crop plants.

Formula (I) provides a general definition of the substituted hetero-fused imidazoles according to the invention. Preferred compounds of the formula (I) are those in which

2158996

R<sup>1</sup> represents hydrogen or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl and alkoxy, each of which has 1 to 8 carbon atoms, or represents phenyl which is optionally monosubstituted or polysubstituted by identical or different substituents, suitable substituents being:

halogen, cyano, nitro in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of which has 1 to 6 carbon atoms, in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkyl-sulphinyl or halogenoalkylsulphonyl each of which has 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, in each case straight-chain or branched alkoxyalkyl, alkoxyalkoxy, alkanoyl, alkoxycarbonyl or alkoximinoalkyl each of which has 1 to 6 carbon atoms in the individual alkyl moieties, or divalent dioxyalkylene having 1 to 5 carbon atoms which is optionally monosubstituted or polysubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 6 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, or phenyl which is optionally monosubstituted or polysubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 6 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms,

R<sup>2</sup> represents hydroxyl, cyano or a straight-chain or branched radical from the series consisting of alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy or dialkoxyphosphonyl, each of which has up to 8 carbon atoms in the individual alkyl or alkenyl or alkinyl moieties and each of these radicals optionally being monosubstituted or polysubstituted by identical or different substituents, suitable substituents in each case being:

fluorine, chlorine, bromine, iodine, straight-chain or branched alkoxy having 1

30

5

10

15

20

to 8 carbon atoms, or aryl having 6 to 10 carbon atoms or heteroaryl having 2 to 9 carbon atoms and 1 to 5 hetero atoms (in particular nitrogen, oxygen and/or sulphur), these aryl or heteroaryl substituents in each case optionally being monosubstituted or polysubstituted by identical or different substituents and suitable aryl or heteroaryl substituents being those mentioned in the case of R<sup>1</sup>,

R<sup>2</sup> furthermore represents amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

5

10

15

20

25

formyl, straight-chain or branched alkyl having 1 to 8 carbon atoms, straightchain or branched alkenyl having 2 to 8 carbon atoms, straight-chain or branched alkylsulphonyl having 1 to 8 carbon atoms, carbamoyl, thiocarbamoyl or sulphamoyl, each of which is monosubstituted or disubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 8 carbon atoms, cycloalkyl, cycloalkylcarbonyl or cycloalkyloxycarbonyl, each of which has 3 to 8 carbon atoms in the cycloalkyl moiety, alkylcarbonyl, alkenylcarbonyl, alkoxycarbonyl, alkenyloxycarbonyl, alkylthio-carbonyl, alkoxythiocarbonyl or alkylthio-thiocarbonyl, each of which has 1 to 8 carbon atoms in the individual straight-chain or branched alkyl moieties, in each case divalent and cyclized alkanediylcarbonyl or alkanediyloxycarbonyl, each of which has 2 to 6 carbon atoms in the alkanediyl moiety, arylalkyl, arylalkylcarbonyl or arylalkyloxycarbonyl, each of which has 6 to 10 carbon atoms in the aryl moiety and 1 to 8 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted in the aryl moiety by identical or different substituents, or aryl, arylcarbonyl or aryloxycarbonyl, each of which has 6 to 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted or polysubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>.

30 R<sup>2</sup> furthermore represents aryl, arylcarbonyl, arylcarbonyl, arylcarbonyloxy or Le A 29 565 - 8 -

arylaminocarbonylaminocarbonyloxy, each of which has 6 to 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted or polysubstituted by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>,

furthermore represents heteroaryl, heteroarylcarbonyl, heteroaryloxycarbonyl, heteroarylcarbonyloxy or heteroarylaminocarbonylaminocarbonyloxy, each of which has 2 to 9 carbon atoms and 1 to 5 identical or different hetero atoms (in particular nitrogen, oxygen and/or sulphur) in the heteroaryl moiety and each of which is optionally monosubstituted or polysubstituted by identical or different substituents, suitable heteroaryl substituents in each case being the aryl substituents mentioned in the case of R<sup>1</sup>.

represents cyano, fluorine, chlorine, bromine, iodine or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of cycloalkyl, alkyl, alkenyl, alkinyl, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy, alkenyloxy, alkenyloxy, alkenyloxy, each of which has up to 8 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties, suitable substituents in each case being: fluorine, chlorine, bromine, iodine, straight-chain or branched alkoxy having 1 to 8 carbon atoms, or aryl having 6 to 10 carbon atoms or heteroaryl having 2 to 9 carbon atoms and 1 to 5 hetero atoms (in particular nitrogen, oxygen and/or sulphur), each of these aryl or heteroaryl radicals optionally being monosubstituted or polysubstituted by identical or different substituents and suitable aryl or heteroaryl substituents being those mentioned in the case of R<sup>1</sup>,

furthermore represents amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

formyl, straight-chain or branched alkyl having 1 to 8 carbon atoms, straight-chain or branched alkenyl having 2 to 8 carbon atoms, straight-chain or branched alkylsulphonyl having 1 to 8 carbon atoms, carbamoyl, thiocarbamoyl

 $\mathbb{R}^3$ 

15

or sulphamoyl, each of which is monosubstituted or disubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 8 carbon atoms, cycloalkyl, cycloalkylcarbonyl or cycloalkyloxycarbonyl, each of which has 3 to 8 carbon atoms in the cycloalkyl moiety, alkylcarbonyl, alkenylcarbonyl, alkoxycarbonyl, alkenyloxycarbonyl, alkylthio-carbonyl, alkoxy-thiocarbonyl or alkylthio-thiocarbonyl, each of which has 1 to 8 carbon atoms in the individual straight-chain or branched alkyl moieties, in each case divalent and cyclized alkanediylcarbonyl or alkanediyloxycarbonyl, each of which has 2 to 6 carbon atoms in the alkanediyl moiety, arylalkyl, arylalkylcarbonyl or arylalkyloxycarbonyl, each of which has 6 to 10 carbon atoms in the aryl moiety and 1 to 8 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted in the aryl moiety by identical or different substituents, or aryl, arylcarbonyl or aryloxycarbonyl, each of which has 6 to 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted or polysubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R1,

- R<sup>3</sup> furthermore represents aryl having in each case 6 to 10 carbon atoms in the aryl moiety which is in each case optionally monosubstituted or polysubstituted by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>.
  - A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> and A<sup>4</sup> in each case represent N(nitrogen), N-CHR<sup>1</sup>R<sup>2</sup> or CX, the heterofused ring having at least one, but not more than two, nitrogen atoms simultaneously and all positional isomers being possible, so that
- 25 CX<sup>1</sup>, CX<sup>2</sup>, CX<sup>3</sup> exist in the case of one nitrogen atom and
  - CX<sup>1</sup> and CX<sup>2</sup> exist in the case of two nitrogen atoms, and, when either A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> or A<sup>4</sup> represent N-CHR<sup>1</sup>R<sup>2</sup>, the imidazole ring exists only in monosubstituted form (R<sup>3</sup>), and

5

10

15

20

P

X1. X2 and X3 in each case independently of one another represent hydrogen, fluorine, chlorine, bromine, iodine, cyano, nitro, in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of which has 1 to 8 carbon atoms, cycloalkyl having 3 to 8 carbon atoms, in each case straightchain or branched halogenoalkyl, halogenoalkylthio, halogenoalkylsulphinyl, halogenoalkylsulphonyl, each of which has 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, or divalent dioxyalkylene having 1 to 5 carbon atoms which is optionally monosubstituted or polysubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, furthermore represent hydroxycarbonyl, in each case straight-chain or branched alkylcarbonyl or alkoxycarbonyl, each of which has 1 to 6 carbon atoms in the alkyl moiety. 15 cycloalkyloxycarbonyl having 3 to 8 carbon atoms in the cycloalkyl moiety, or amino or aminocarbonyl, each of which is optionally monosubstituted or polysubstituted by identical or different substituents, suitable amino substituents in each case being:

> in each case straight-chain or branched alkyl having 1 to 6 carbon atoms, halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 halogen atoms, alkoxyalkyl or alkylcarbonyl, each of which has 1 to 6 carbon atoms in the individual alkyl moieties, or arylcarbonyl, arylsulphonyl, arylaminocarbonyl or arylmethylsulphonyl, each of which has 6 to 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted or polysubstituted in the arvl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R1;

X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> furthermore represent aryl, aryloxy, arylthio, arylsulphinyl, arylsulphonyl, arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylthiomethylsulphonyl or arylazo, each of which has 6 to 10 carbon atoms in the aryl mojety and each of which is optionally monosubstituted or polysubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each

30

5

10

20

case being those mentioned in the case of R1, and

where at least one of the substituents X1, X2 or X3 represents in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogeno-alkylsulphinyl, halogenoalkylsulphonyl, each of which has 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, or represents straight-chain or branched alkylsulphonyl having 1 to 6 carbon atoms, or divalent dioxyalkylene having 1 to 5 carbon atoms which is optionally monosubstituted or polysubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, furthermore represents hydroxycarbonyl, in each case straight-chain or branched alkylcarbonyl or alkoxycarbonyl, each of which has 1 to 6 carbon atoms in the alkyl moiety, cycloalkyloxycarbonyl having 3 to 8 carbon atoms in the cycloalkyl moiety, or amino or aminocarbonyl, each of which is optionally monosubstitued or polysubstituted by identical or different substituents, suitable amino substituents in each case being:

in each case straight-chain or branched alkyl having 1 to 6 carbon atoms, halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 halogen atoms, alkoxyalkyl or alkylcarbonyl, each of which has 1 to 6 carbon atoms in the individual alkyl moieties, or arylcarbonyl, arylsulphonyl, arylaminocarbonyl or arylmethylsulphonyl, each of which has 6 to 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted or polysubstituted by identical or different substituents in the aryl moiety, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>,

X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> furthermore represent aryl, arylthio, arylsulphinyl, arylsulphonyl, arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylthiomethylsulphonyl or arylazo, each of which has 6 to 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted or polysubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being

30

5

10

15

20

those mentioned in the case of R1.

5

10

15

20

25

Particularly preferred compounds of the formula (I) are those in which

R¹ represents hydrogen, or a straight-chain or branched radical from the series consisting of alkyl and alkoxy, each of which has 1 to 6 carbon atoms and each of which is unsubstituted or substituted, or represents phenyl which is optionally monosubstituted to trisubstituted by identical or different substituents, suitable substituents being:

fluorine, chlorine, bromine, iodine, cyano, nitro, in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of which has 1 to 4 carbon atoms, in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl or halogenoalkylsulphonyl, each of which has 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, in each case straight-chain or branched alkoxyalkyl, alkoxyalkoxy, alkanoyl, alkoxycarbonyl or alkoximinoalkyl, each of which has 1 to 4 carbon atoms in the individual alkyl moieties, divalent dioxyalkylene having 1 to 4 carbon atoms which is optionally monosubstituted to hexasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, or phenyl which is optionally monosubstituted to pentasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, halogen in each case representing fluorine, chlorine, bromine or iodine,

R<sup>2</sup> represents hydroxyl, cyano, or a straight-chain or branched radical from the series consisting of alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy and dialkoxyphosphonyl, each of which has up to 6 carbon atoms in the individual

alkyl, alkenyl or alkinyl moieties and each of which is optionally monosubstituted to pentasubstituted by identical or different substituents from the series consisting of fluorine, chlorine, bromine and iodine, or represents alkyl, alkenyl or alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy or dialkoxyphosphonyl, each of which has up to 6 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties and each of which is optionally monosubstituted to trisubstituted by identical or different substituents, suitable substituents in each case being:

straight-chain or branched alkoxy having 1 to 6 carbon atoms, or aryl having 6 or 10 carbon atoms or heteroaryl having 2 to 9 carbon atoms and 1 to 4 hetero atoms (in particular nitrogen, oxygen and/or sulphur), each of these aryl or heteroaryl radicals optionally being monosubstituted to trisubstituted by identical or different substituents and suitable aryl or heteroaryl substituents being those mentioned in the case of R<sup>1</sup>,

R<sup>2</sup> furthermore represents amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

formyl, straight-chain or branched alkyl having 1 to 6 carbon atoms, straight-chain or branched alkenyl having 2 to 6 carbon atoms, straight-chain or branched alkylsulphonyl having 1 to 6 carbon atoms, carbamoyl, thiocarbamoyl or sulphamoyl, each of which is optionally monosubstituted or disubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 6 carbon atoms, or cycloalkyl, cycloalkylcarbonyl or cycloalkyloxycarbonyl, each of which has 3 to 7 carbon atoms in the cycloalkyl moiety, alkylcarbonyl, alkenylcarbonyl, alkoxycarbonyl, alkenyloxycarbonyl, alkylthio-carbonyl, alkoxy-thiocarbonyl or alkylthio-thiocarbonyl, each of which has 1 to 6 carbon atoms in the individual straight-chain or branched alkyl moieties, in each case divalent and cyclized alkanediylcarbonyl or alkanediyloxycarbonyl, each of which has 2 to 5 carbon atoms in the alkanediyl moiety, arylalkyl, arylalkylcarbonyl or arylalkyloxycarbonyl, each of which has 6 or 10 carbon

30

25

5

10

15

atoms in the aryl moiety and 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted to trisubstituted in the aryl moiety by identical or different substituents, or aryl, arylcarbonyl or aryloxycarbonyl, each of which has 6 or 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted to trisubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>,

R<sup>2</sup> furthermore represents aryl, arylcarbonyl, arylcarbonyl, arylcarbonyloxy or arylaminocarbonylaminocarbonyloxy, each of which has 6 or 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted to pentasubstituted by identical or different substituents, suitable aryl substituents in each case being those mentioned under R<sup>1</sup>,

furthermore represents heteroaryl, heteroarylcarbonyl, heteroarylcarbonyloxy or heteroarylaminocarbonylaminocarbonyloxy, each of which has 2 to 9 carbon atoms and 1 to 4 identical or different hetero atoms (in particular nitrogen, oxygen and/or sulphur) in the heteroaryl moiety and each of which is optionally monosubstituted to pentasubstituted by identical or different substituents, suitable heteroaryl substituents in each case being the aryl substituents mentioned in the case of R<sup>1</sup>,

represents cyano, fluorine, chlorine, bromine, iodine, or a straight-chain or branched radical from the series consisting of alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl and alkylcarbonyloxy, each of which has up to 6 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties and which is optionally monosubstituted to pentasubstituted by identical or different substituents from the series consisting of fluorine, chlorine, bromine and iodine, or represents cycloalkyl, alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy or dialkoxyphosphonyl, each of which has up to 6 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties and each of which is optionally monosubstituted to trisubstituted by identical or different

30

5

10

15

20

25

 $R^2$ 

 $\mathbb{R}^3$ 

substituents, suitable substituents in each case being:

fluorine, chlorine, bromine, iodine, straight-chain or branched alkoxy having 1 to 6 carbon atoms, or aryl having 6 or 10 carbon atoms or heteroaryl having 2 to 9 carbon atoms and 1 to 4 hetero atoms (in particular nitrogen, oxygen and/or sulphur), each of these aryl or heteroaryl radicals optionally being monosubstituted to trisubstituted by identical or different substituents, suitable aryl or heteroaryl substituents being those mentioned in the case of R<sup>1</sup>,

R³ furthermore represents amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

formyl, straight-chain or branched alkyl having 1 to 6 carbon atoms, straightchain or branched alkenyl having 2 to 6 carbon atoms, straight-chain or branched alkylsulphonyl having 1 to 6 carbon atoms, carbamoyl, thiocarbamoyl or sulphamoyl, each of which is optionally monosubstituted or disubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 6 carbon atoms, or cycloalkyl, cycloalkylcarbonyl, or cycloalkyloxycarbonyl, each of which has 3 to 7 carbon atoms in the cycloalkyl moiety, alkylcarbonyl, alkenylcarbonyl, alkoxycarbonyl, alkylthio-carbonyl, alkylthio-carbonyl, alkoxy-thiocarbonyl or alkylthio-thiocarbonyl, each of which has 1 to 6 carbon atoms in the individual straight-chain or branched alkyl moieties, in each case divalent and cyclized alkanediylcarbonyl or alkanediyloxycarbonyl, each of which has 2 to 5 carbon atoms in the alkanediyl moiety, or arylalkyl, arylalkylcarbonyl or arylalkyloxycarbonyl, each of which has 6 or 10 carbon atoms in the aryl moiety and 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted to trisubstituted by identical or different substituents in the aryl moiety, or aryl, arylcarbonyl or aryloxycarbonyl, each of which has 6 or 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted to trisubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R1,

30

5

10

15

20

- R³ furthermore represents aryl having in each case 6 or 10 carbon atoms in the aryl moiety which is in each case optionally monosubstituted to pentasubstituted by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R¹,
- 5 A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> and A<sup>4</sup> in each case represent N(nitrogen), N-CHR<sup>1</sup>R<sup>2</sup> or CX, the heterofused ring having at least one, but not more than two, nitrogen atoms simultaneously and all positional isomers being possible, so that
  - CX1, CX2, CX3 exist in the case of one nitrogen atom and
- CX<sup>1</sup> and CX<sup>2</sup> exist in the case of two nitrogen atoms, and, when either A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> or

  A<sup>4</sup> represent N-CHR<sup>1</sup>R<sup>2</sup>, the imidazole ring exists only in monosubstituted form
  (R<sup>3</sup>), and
- X1, X2 and X3 in each case independently of one another represent hydrogen, fluorine, chlorine, bromine, cyano, nitro, in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of which has 1 to 6 15 carbon atoms, cycloalkyl having 3 to 7 carbon atoms, in each case straightchain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl, halogenoalkylsulphonyl, each of which has 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, or divalent dioxyalkylene having 1 to 4 carbon atoms which is optionally monosubstituted 20 to hexasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, furthermore represent hydroxycarbonyl, in each case straight-chain or branched alkylcarbonyl or 25 alkoxycarbonyl, each of which has 1 to 4 carbon atoms in the alkyl moiety, cycloalkyloxycarbonyl having 3 to 7 carbon atoms in the cycloalkyl moiety, or amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable amino substituents in each case being:

in each case straight-chain or branched alkyl having 1 to 4 carbon atoms, halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 halogen atoms, alkoxyalkyl or alkylcarbonyl, each of which has 1 to 4 carbon atoms in the individual alkyl moieties, or arylcarbonyl, arylsulphonyl, arylaminocarbonyl or arylmethylsulphonyl, each of which has 6 or 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted to pentasubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>;

X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> furthermore represent aryl, aryloxy, arylthio, arylsulphinyl, arylsulphonyl, arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylthiomethylsulphonyl or arylazo, each of which has 6 or 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted to pentasubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>, and

where at least one of the substituents X1, X2 and X3 represents in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl, halogenoalkylsulphonyl, each of which has 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, straight-chain or branched alkylsulphonyl having 1 to 4 carbon atoms, or divalent dioxyalkylene having 1 to 4 carbon atoms which is optionally monosubstituted to hexasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, furthermore represents hydroxycarbonyl, in each case straight-chain or branched alkylcarbonyl or alkoxycarbonyl, each of which has 1 to 4 carbon atoms in the alkyl moiety, cycloalkyloxycarbonyl having 3 to 7 carbon atoms in the cycloalkyl moiety, or amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable amino substituents in each case being:

5

15

20

25

in each case straight-chain or branched alkyl having 1 to 4 carbon atoms, halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 halogen atoms, alkoxyalkyl or alkylcarbonyl, each of which has 1 to 4 carbon atoms in the individual alkyl moieties, or arylcarbonyl, arylsulphonyl arylaminocarbonyl, or arylmethylsulphonyl, each of which has 6 or 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted to pentasubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>;

- X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> furthermore represent aryl, arylthio, arylsulphinyl, arylsulphonyl, arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylthiomethylsulphonyl or arylazo, each of which has 6 or 10 carbon atoms in the aryl moiety, such as phenyl or naphthyl, and each of which is optionally monosubstituted to pentasubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>.
- 15 Very particularly preferred compounds of the formula (I) are those in which
  - R<sup>1</sup> represents hydrogen or a straight-chain or branched radical from the series consisting of alkyl and alkoxy, each of which has 1 to 4 carbon atoms and each of which is unsubstituted or substituted, or represents phenyl which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents being:

fluorine, chlorine, bromine, cyano, nitro, in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of which has 1 to 3 carbon atoms, in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl or halogenoalkylsulphonyl, each of which has 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, in each case straight-chain or branched alkoxyalkyl, alkoxyalkoxy, alkanoyl, alkoxycarbonyl or alkoximinoalkyl, each of which has 1 to 3 carbon atoms in the individual alkyl moieties, divalent dioxyalkylene having 1 to 3 carbon atoms which is optionally monosubstituted to

25

20

tetrasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, or phenyl which is optionally monosubstituted to trisubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 3 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, halogen in each case representing fluorine, chlorine or bromine,

5

25

represents hydroxyl, cyano or a straight-chain or branched radical from the 10  $\mathbb{R}^2$ series consisting of alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy and dialkoxyphosphonyl, each of which has up to 4 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties and each of which is optionally monosubstituted to tetrasubstituted by identical or different substituents from the series consisting 15 of fluorine, chlorine and bromine, or represents alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy or dialkoxyphosphoryl, each of which has up to 4 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties, and each of which is 20 optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

straight-chain or branched alkoxy having 1 to 3 carbon atoms or phenyl which is optionally monosubstituted or disubstituted by identical or different substituents, suitable phenyl substituents being those mentioned in the case of  $R^1$ ,

R<sup>2</sup> furthermore represents amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

formyl, straight-chain or branched alkyl having 1 to 4 carbon atoms,

Le A 29 565

- 20 -

straight-chain or branched alkenyl having 2 to 4 carbon atoms, straight-chain or branched alkylsulphonyl having 1 to 4 carbon atoms, carbamoyl, thiocarbamoyl or sulphamoyl, each of which is optionally monosubstituted or disubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 4 carbon atoms, cycloalkyl, cycloalkylcarbonyl or cycloalkyloxycarbonyl, each of which has 3 to 6 carbon atoms in the cycloalkyl moiety, alkylcarbonyl, alkenylcarbonyl, alkoxycarbonyl, alkenyloxycarbonyl, alkylthio-carbonyl, alkoxy-thiocarbonyl or alkylthio-thiocarbonyl, each of which has 1 to 4 carbon atoms in the individual straight-chain or branched alkyl moieties, in each case divalent and cyclized alkanediylcarbonyl or alkanediyloxycarbonyl, each of which has 2 to 4 carbon atoms in the alkanediyl moiety, phenylalkyl, phenylalkylcarbonyl or phenylalkyloxycarbonyl, each of which has 1 to 4 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or disubstituted in the phenyl moiety by identical or different substituents, or phenyl, phenylcarbonyl or phenyloxycarbonyl, each of which is optionally monosubstituted or disubstituted in the phenyl moiety by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of R<sup>1</sup>,

 $R^2$ phenylcarbonyl, furthermore represents phenyl, phenyloxycarbonyl, 20 phenylcarbonyloxy or phenylaminocarbonylaminocarbonyloxy, each of which is optionally monosubstituted to trisubstituted by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of  $R^{1}$ ,

 $R^2$ furthermore represents heteroaryl, heteroarylcarbonyl, heteroaryloxycarbonyl, 25 heteroarylcarbonyloxy or heteroarylaminocarbonylaminocarbonyloxy, each of which have 2 to 9 carbon atoms and 1 to 3 identical or different hetero atoms (in particular nitrogen, oxygen and/or sulphur) in the heteroaryl moiety and each of which is optionally monosubstituted to trisubstituted by identical or different substituents, suitable heteroaryl substituents in each case being the phenyl substituents mentioned in the case of R<sup>1</sup>,

30

5

10

 $\mathbb{R}^3$ represents cyano, fluorine, chlorine, bromine, or a straight-chain or branched radical from the series consisting of alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl and alkylcarbonyloxy, each of which has up to 4 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties and each of which is optionally monosubstituted to trisubstituted by identical or different substituents from the series consisting of fluorine, chlorine and bromine, or represents alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy or dialkoxyphosphoryl, each of which has up to 4 carbon atoms in the individual 10 alkyl, alkenyl or alkinyl moieties and each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

> straight-chain or branched alkoxy having 1 to 3 carbon atoms or phenyl which is optionally monosubstituted or disubstituted by identical or different substituents, suitable phenyl substituents being those mentioned in the case of R1,

 $R^3$ furthermore represents amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

formyl, straight-chain or branched alkyl having 1 to 4 carbon atoms, straightchain or branched alkenyl having 2 to 4 carbon atoms, straight-chain or branched alkylsulphonyl having 1 to 4 carbon atoms, in each case optionally monosubstituted or disubstituted (identically or differently by straight-chain or branched alkyl having 1 to 4 carbon atoms) carbamoyl, thiocarbamoyl or sulphamoyl, cycloalkyl, cycloalkylcarbonyl, or cycloalkyloxycarbonyl, each of which has 3 to 6 carbon atoms in the cycloalkyl moiety, alkylcarbonyl, alkenylcarbonyl, alkoxycarbonyl, alkenyloxycarbonyl, alkylthio-carbonyl, alkoxy-thiocarbonyl or alkylthio-thiocarbonyl, each of which has 1 to 4 carbon atoms in the individual straight-chain or branched alkyl mojeties, in each case divalent and cyclized alkanediylcarbonyl or alkanediyloxycarbonyl, each of

5

15

20

which has 2 to 4 carbon atoms in the alkanediyl moiety, phenylalkyl, phenylalkylcarbonyl or phenylalkyloxycarbonyl, each of which has 1 to 4 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or disubstituted in the phenyl moiety by identical or different substituents, or phenyl, phenylcarbonyl or phenyloxycarbonyl, each of which is optionally monosubstituted or disubstituted in the phenyl moiety by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of R<sup>1</sup>,

furthermore represents phenyl which is optionally monosubstituted to trisubstituted by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of R<sup>1</sup>,

A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> and A<sup>4</sup> in each case represent N(nitrogen), N-CHR<sup>1</sup>R<sup>2</sup> or CX, the heterofused ring having at least one, but not more than two, nitrogen atoms simultaneously and all positional isomers being possible, so that

15 CX<sup>1</sup>, CX<sup>2</sup>, CX<sup>3</sup> exist in the case of one nitrogen atom and

CX<sup>1</sup> and CX<sup>2</sup> exist in the case of two nitrogen atoms, and, when either A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> or A<sup>4</sup> represent N-CHR<sup>1</sup>R<sup>2</sup>, the imidazole ring exists only in monosubstituted form (R<sup>3</sup>), and

X¹, X² and X³ independently of one another in each case represent hydrogen, chlorine, bromine, cyano, nitro, in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of which has 1 to 4 carbon atoms, cycloalkyl having 3, 5 or 6 carbon atoms, in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl, halogenoalkylsulphonyl, each of which has 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, or represent divalent dioxyalkylene having 1 to 3 carbon atoms which is optionally monosubstituted to tetrasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 3 carbon

atoms and straight-chain or branched halogenoalkyl having 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, furthermore represent hydroxycarbonyl, in each case straight-chain or branched alkylcarbonyl or alkoxycarbonyl, each of which has 1 to 3 carbon atoms in the alkyl moiety, cycloalkyloxycarbonyl having 3, 5 or 6 carbon atoms in the cycloalkyl moiety, or amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable amino substituents in each case being:

10

5

in each case straight-chain or branched alkyl having 1 to 3 carbon atoms, halogenoalkyl having 1 to 3 carbon atoms and 1 to 7 halogen atoms, alkoxyalkyl or alkylcarbonyl, each of which has 1 to 3 carbon atoms in the individual alkyl moieties, or phenylcarbonyl, phenylsulphonyl, phenylaminocarbonyl or phenylmethylsulphonyl, each of which is optionally monosubstituted to trisubstituted in the phenyl moiety by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of R<sup>1</sup>;

15

X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> furthermore represent phenyl, phenyloxy, phenylthio, phenylsulphinyl, phenylsulphonyl, phenylsulphonyloxy, phenylcarbonyl, phenyloxycarbonyl, phenylthiomethylsulphonyl or phenylazo, each of which is optionally monosubstituted to trisubstituted in the phenyl moiety by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of R<sup>1</sup>, and

20

where at least one of the substituents X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> represents in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl, halogenoalkylsulphonyl, each of which has 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, or represents straight-chain or branched alkylsulphonyl having 1 to 3 carbon atoms, or represents divalent dioxyalkylene having 1 to 3 carbon atoms which is optionally monosubstituted to tetrasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched

25

alkyl having 1 to 3 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, furthermore represents hydroxycarbonyl, in each case straight-chain or branched alkylcarbonyl or alkoxycarbonyl, each of which has 1 to 3 carbon atoms in the alkyl moiety, cycloalkyloxycarbonyl having 3, 5 or 6 carbon atoms in the cycloalkyl moiety, or amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable amino substituents in each case being:

10

5

in each case straight-chain or branched alkyl having 1 to 3 carbon atoms, halogenoalkyl having 1 to 3 carbon atoms and 1 to 7 halogen atoms, alkoxyalkyl or alkylcarbonyl, each of which has 1 to 3 carbon atoms in the individual alkyl moieties, or phenylcarbonyl, phenylsulphonyl, phenylaminocarbonyl or phenylmethylsulphonyl, each of which is optionally monosubstituted to trisubstituted in the phenyl moiety by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of R<sup>1</sup>;

15

X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> furthermore represent phenyl, phenylthio, phenylsulphinyl, phenylsulphonyl, phenylsulphonyloxy, phenylcarbonyl, phenylsulphonyl or phenylazo, each of which is optionally monosubstituted to trisubstituted in the phenyl moiety by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of R<sup>1</sup>.

20

Substituted hetero-fused imidazoles of the general formula (I)

$$\begin{array}{c|c}
A & A \\
\downarrow_{2} & N \\
A & N \\
\hline
CH-R^{1} \\
\downarrow_{2} \\
R
\end{array}$$
(I)

which may be mentioned individually in addition to the compounds mentioned in the preparation examples are those which follow:

Table I

<b>x</b> <sup>1</sup>	<b>X</b> <sup>2</sup>	х3	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>
Н	н	н	Н	OEt	CF <sub>3</sub>
Н	н	H	H	OPr	CF <sub>3</sub>
H	н	H	H	OCH≖CH	CF <sub>3</sub>
H	н	H	H	OiPr	CF <sub>3</sub>
H	Н	H	H	OnBu	CF <sub>3</sub>
H	H	H	H	OiBu	CF <sub>3</sub>
H	H	H	H ·	OtBu	CF <sub>3</sub>
H	H	H	H	Osec.Bu	CF <sub>3</sub>
H	H	H	H	OCH <sub>2</sub> CH <sub>2</sub> OMe	CF <sub>3</sub>
H	H	H	H	OCH <sub>2</sub> CH <sub>2</sub> OEt	CF <sub>3</sub>
H	H	Н	H	N-COOE:	CF <sub>3</sub>
Н	Н	н	н	Me N-COOEs	CF <sub>3</sub>
Н	н	Н	н	Et N-COOEt	CF <sub>3</sub>
н	Н	Н	Н	Pr N-COOE	CF <sub>3</sub>
Н	н	Н	Н	N-COOE:	CF <sub>3</sub>
Н	н	н	Н	tBu N-COOEt	CF <sub>3</sub>
Н	. н	н	Н	nPr N-COOEs	CF <sub>3</sub>
Н	н	Н	н	iPr N-COOEs	CF <sub>3</sub>

<b>X</b> 1	X <sup>2</sup>	X <sup>3</sup>	R <sup>1</sup>	R <sup>2</sup>	R <sup>3</sup>
Н	Н	Н	н	O "C	CF <sub>3</sub>
Н	Н	н	Н	0 "C"	CF <sub>3</sub>
H	Br	н	н	<u> </u>	i i
Br	н	н	н		
H	Br	н	н		
H	CI	н	Н		
Cl	H	н	н		
H	H	Cl	н		
F	Н	н	Н		
H	F	н	Н		
H	Н	F	H		
H	CF <sub>3</sub>	H	H		
CF <sub>3</sub>	н	H	H		
H	н	CF <sub>3</sub>	H		
H	OCF <sub>3</sub>	Н	H		
H	SCF <sub>3</sub>	Н	H		
H	NO <sub>2</sub>	H	H		
H	CHF <sub>2</sub>	н	H		
H	OCHF <sub>2</sub>	Н	H		
H	Н	Н	Н	$CH = CH_2$	CF <sub>3</sub>
H	Н	Н	Н	C=CH	CF <sub>3</sub>
H	Н	Н	H	сосн3	CF <sub>3</sub>
H	H	н	H	H3CCONH-	CF <sub>3</sub>
H	н	н	H	(H <sub>3</sub> ) <sub>2</sub> CCONH-	CF <sub>3</sub>

All the examples also apply to  $R^3 = CHF_2$ ,  $R^3 = C_2F_5$ ,  $R^3 = C_3F_7$ , and, additionally, the  $R^2$  and  $R^3$  radicals can be varied, as shown in the Table, for each  $X^1$  to  $X^3$  pattern.

The substituent variations shown in the above table can also be given for the other isomeric pyridines:

5 This variation, limited to  $X^1$  and  $X^2$ , also applies analogously to the pyrimidinoimidazoles

#### the pyridazines

and the pyrazines

If, for example, the pyrimidinoimidazole (1) and chloromethyl ethyl ester are used as starting compounds, the course of the reaction of the process according to the invention can be represented by the following equation:

3 
$$\longrightarrow$$
 CF<sub>3</sub> + 3 CICH<sub>2</sub>-O-C<sub>2</sub>H<sub>5</sub>  $\xrightarrow{base}$  -HCI

(1)

 $\longrightarrow$  CF<sub>3</sub> +  $\longrightarrow$  CF<sub>3</sub> +  $\longrightarrow$  CF<sub>3</sub>  $\longrightarrow$  CH<sub>2</sub>OC<sub>2</sub>H<sub>5</sub>  $\xrightarrow{cH_2OC_2H_5}$   $\xrightarrow{cH_2OC_2H_5}$   $\xrightarrow{cH_2OC_2H_5}$ 

Formula (II) provides a general definition of the hetero-fused imidazoles required as starting materials for carrying out the process according to the invention. In this formula (II), A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup>, A<sup>4</sup> and R<sup>3</sup> preferably represent those radicals which have already been mentioned in connection with the description of the compound of the formula (I) according to the invention as being preferred for these substituents.

The 1H-hetero-fused imidazoles of the formula (II) are known or can be obtained in analogy to known processes (GB 1 114 199; JP 62 294 683; J. Heterocyl. Chem. 18 (2), 303-7; EP 297 661; J. Med. Chem. 33 (8), 2231-9).

Formula (III) provides a general definition of the compounds furthermore required as educts for carrying out the process according to the invention. In this formula (III), R<sup>1</sup> and R<sup>2</sup> preferably represent those radicals which have already been mentioned in connection with the substances of the formula (I) according to the invention as being preferred for the substituents.

M represents a leaving radical customary in alkylating agents, preferably halogen, arylsulphonate, arylalkylsuphonate, alkylsulphonate, alkylsulphonate, arylcarbonyloxy, particularly preferably chlorine, bromine, iodine, C<sub>1-8</sub>-alkylsulphonate, tolylsulphonate, phenylsulphonate, C<sub>1-8</sub>-alkylcarbonyloxy, or benzoyl, and particularly preferably chlorine, bromine, C<sub>1</sub>-C<sub>2</sub>-alkylsulphonate, phenylsulphonate, tolylsulphonate, C<sub>1</sub>-C<sub>3</sub>-alkylcarbonyloxy or benzoyl.

5

10

The compounds of the formula (III) are known or can be obtained in analogy to known processes (cf., for example, DE 20 40 175; DE 21 19 518; Synthesis 1973, 703).

Suitable diluents for carrying out the process according to the invention are inert organic solvents. These include, in particular, aliphatic, alicyclic or aromatic, optionally halogenated hydrocarbons, such as, for example, benzine, benzene, toluene, xylene, chlorobenzene, dichlorobenzene, petroleum ether, hexane, cyclohexane, dichloromethane, chloroform or carbon tetrachloride; ethers, such as diethyl ether, diisopropyl ether, dioxane, tetrahydrofuran, ethylene glycol dimethyl ether or ethylene glycol diethyl ether; ketones, such as acetone, butanone or methyl isobutyl ketone; nitriles, such as acetonitrile, propionitrile or benzonitrile; amides, such as N,N-dimethylformamide, N,N-dimethylacetamide, N-methylformanilide N-methylpyrrolidone or hexamethylphosphoric triamide; esters, such as methyl acetate or ethyl acetate, or bases such as pyridine, or organic acids such as formic acid or acetic acid.

The process according to the invention is preferably carried out in the presence of a suitable reaction auxiliary. Suitable reaction auxiliaries are all customary inorganic or organic bases. These include, for example, the hydrides, hydroxides, amides, alcoholates, acetates, carbonates or hydrogen carbonates of alkaline earth metals or alkali metals, such as, for example, sodium hydride, sodium amide, lithium diethylamide, sodium methylate, sodium ethylate, potassium tert-butylate, sodium hydroxide, potassium hydroxide, ammonium hydroxide, sodium acetate, potassium acetate, calcium acetate, ammonium acetate, sodium carbonate, potassium carbonate, potassium hydrogen carbonate, sodium hydrogen carbonate or ammonium carbonate, organolithium compounds, such as n-butyllithium, and also tertiary amines, such as trimethylamine, triethylamine, tributylamine, di-isopropyl-ethylamine, tetramethylguanidine, N,N-dimethylaniline, pyridine, piperidine, N-methylpiperidine, N,N-dimethylaminopyridine, diazabicyclooctane (DABCO), diazabicyclononene (DBN) or diazabicycloundecene (DBU).

In those cases where A in formula (III) represents an alcohol, alkanoyloxy or alkoxy group, suitable reaction auxiliaries also include organic or inorganic acids, such as, for example, sulphuric acid, hydrochloric acid, p-toluenesulphonic acid,

5

10

15

20

25

perfluorobutanesulphonic acid or strongly acidic ion exchangers.

If appropriate, the process according to the invention can also be carried out in a twophase system, such as, for example, water/toluene or water/dichloromethane, if appropriate in the presence of a suitable phase transfer catalyst. Examples of such catalysts which may be mentioned are: tetrabutylammonium iodide. tetrabutylammonium chloride, tetrabutylammonium bromide, tributylmethylphosphonium bromide, trimethyl-C<sub>1</sub>/C<sub>15</sub>-alkylammonium chloride, trimethyl-C<sub>13</sub>/C<sub>15</sub>-alkylammonium bromide, dibenzyl-dimethyl-ammonium methylsulphate, dimethyl-C<sub>12</sub>/C<sub>14</sub>-alkylbenzylammonium chloride, dimethyl-C<sub>12</sub>/C<sub>14</sub>alkylbenzylammonium bromide, tetrabutylammonium hydroxide, triethylbenzylammonium chloride, methyltrioctylammonium chloride, trimethylbenzylammonium chloride, 15-krone-5, 18-krone-6 tris-[2-(2methoxyethoxy)-ethyl]-amine.

When carrying out the process according to the invention, the reaction temperatures can be varied within a substantial range. In general, the process is carried out at temperatures between -70°C and +200°C, preferably at temperatures between 0°C and 130°C.

The process according to the invention is conventionally carried out under atmospheric pressure. However, it can also be carried out under elevated or reduced pressure.

To carry out the process according to the invention, 1.0 to 5.0 mol, preferably 1.0 to 2.5 mol, of the compound of the formula (III) and, if appropriate, 0.01 to 5.0 mol, preferably 1.0 to 3.0 mol, of reaction auxiliary are generally employed per mole of 1H-hetero-fused imidazole of the formula (II).

In a particular embodiment, it is also possible to first silylate the 1H-hetero-fused imidazoles of the formula (II) in a preceding reaction step with the aid of customary silylation methods, for example with hexamethyldisilazane or trimethylsilyl chloride, at temperatures between -20°C and +50°C, if appropriate in the presence of a suitable catalyst, such as, for example, sulphuric acid, trifluoroacetic acid, ammonium sulphate, imidazole or saccharin, and to react the resulting hetero-fused

5

10

15

20

1-trimethylsilylimidazoles in a subsequent second step with alkylating agents of the formula (II) in accordance with the process according to the invention. In this case, it is advantageous to add tin tetrachloride as a catalyst for the alkylation reaction (cf., for example, Chem. Heterocycl. Comp. USSR 24, 514 [1988])

The reaction is carried out and the reaction products are worked up and isolated by known methods (cf. in this context also the preparation examples).

The end products of the formula (I) are purified with the aid of customary processes, for example by column chromatography or by recrystallization.

They are characterized with the aid of the melting point or, in the case of compounds
which do not crystallize - in particular in the case of regio isomer mixtures -, with the
aid of proton nuclear resonance spectroscopy (<sup>1</sup>H NMR).

The active compounds according to the invention are suitable for combating animal pests, preferably arthropods and nematodes, in particular insects and arachnida, which are encountered in agriculture, in forestry, in the protection of stored products and of materials, and in the hygiene field. They are active against normally sensitive and resistant species and against all or some stages of development. The abovementioned pests include:

From the order of the Isopoda, for example, Oniscus asellus, Armadillidium vulgare and Porcellio scaber.

From the order of the Diplopoda, for example, Blaniulus guttulatus

From the order of the Chilopoda, for example, Geophilus carpophagus and Scutigera spec.

From the order of the Symphyla, for example, Scutigerella immaculata.

From the order of the Thysanura, for example, Lepisma saccharina.

From the order of the Collembola, for example, Onychiurus armatus.

From the order of the Orthoptera, for example, Blatta orientalis, Periplaneta americana,
Leucophaea maderae, Blattella germanica, Acheta domesticus, Gryllotalpa spp., Locusta

## 2158996

migratoria migratorioides. Melanoplus differentialis and Schistocerca gregaria.

From the order of the Dermaptera, for example, Forficula auricularia.

From the order of the Isoptera, for example, Reticulitermes spp..

From the order of the Anoplura, for example, Phylloxera vastatrix, Pemphigus spp.,

Pediculus humanus corporis, Haematopinus spp. and Linognathus spp.. 5

15

20

25

Le A 29 565

From the order of the Mallophaga, for example, Trichodectes spp. and Damalinea spp. From the order of the Thysanoptera, for example, Hercinothrips femoralis and Thrips tabaci.

From the order of the Heteroptera, for example, Eurygaster spp., Dysdercus intermedius, Piesma quadrata, Cimex lectularius, Rhodnius prolixus and Triatoma spp. 10

From the order of the Homoptera, for example, Aleurodes brassicae, Bemisia tabaci, Trialeurodes vaporariorum, Aphis gossypii, Brevicoryne brassicae, Cryptomyzus ribis, Aphis fabae, Doralis pomi, Eriosoma lanigerum, Hyalopterus arundinis, Macrosiphum avenae, Myzus spp., Phorodon humuli, Rhopalosiphum padi, Empoasca spp., Euscelis bilobatus, Nephotettix cincticeps, Lecanium corni, Saissetia oleae, Laodelphax striatellus, Nilaparvata lugens, Aonidiella aurantii, Aspidiotus hederae, Pseudococcus spp. and Psylla spp.

From the order of the Lepidoptera, for example, Pectinophora gossypiella, Bupalus piniarius, Cheimatobia brumata, Lithocolletis blancardella, Hyponomeuta padella, Plutella maculipennis, Malacosoma neustria, Euproctis chrysorrhoea, Lymantria spp. Bucculatrix thurberiella, Phyllocnistis citrella, Agrotis spp., Euxoa spp., Feltia spp., Earias insulana, Heliothis spp., Spodoptera exigua, Mamestra brassicae, Panolis flammea, Prodenia litura, Spodoptera spp., Trichoplusia ni, Carpocapsa pomonella, Pieris spp., Chilo spp., Pyrausta nubilalis, Ephestia kuehniella, Galleria mellonella, Tineola bisselliella, Tinea pellionella, Hofmannophila pseudospretella, Cacoecia podana, Capua reticulana, Choristoneura fumiferana, Clysia ambiguella, Homona magnanima and Tortrix viridana.

From the order of the Coleoptera, for example, Anobium punctatum, Rhizopertha - 35 -

dominica, Acanthoscelides obtectus, Acanthoscelides obtectus, Hylotrupes bajulus, Agelastica alni, Leptinotarsa decemlineata, Phaedon cochleariae, Diabrotica spp., Psylliodes chrysocephala, Epilachna varive stis, Atomaria spp., Oryzaephilus surinamensis, Antho nomus spp., Sitophilus spp., Otiorrhynchus sulcatus, Cosmopolites sordidus, Ceuthorrhynchus assimilis, Hypera postica, Dermestes spp., Trogoderma spp., Anthrenus spp., Attagenus spp., Lyctus spp., Meligethes aeneus, Ptinus spp., Niptus hololeucus, Gibbium psylloides, Tribolium spp., Tenebrio molitor, Agriotes spp., Cono derus spp., Melolontha melolontha, Amphimallon solsti tialis and Costelytra zealandica.

From the order of the Hymenoptera, for example, Diprion spp., Hoplocampa spp.,

10 Lasius spp., Monomorium pharaonis and Vespa spp.

From the order of the Diptera, for example, Aedes spp., Anopheles spp., Culex spp., Drosophila melanogaster, Musca spp., Fannia spp., Calliphora erythrocephala, Lucilia spp., Chrysomyia spp., Cuterebra spp., Gastrophilus spp., Hyppobosca spp., Stomoxys spp., Oestrus spp., Hypoderma spp., Tabanus spp., Tannia spp., Bibio hortulanus, Oscinella frit, Phorbia spp., Pegomyia hyoscyami, Ceratitis capitata, Dacus oleae and Tipula paludosa.

From the order of the Siphonaptera, for example, Xenopsylla cheopis and Ceratophyllus spp..

From the order of the Arachnida, for example, Scorpio maurus and Latrodectus mactans.

From the order of the Acarina, for example, Acarus siro, Argas spp., Omithodoros spp., Dermanyssus gallinae, Eriophyes ribis, Phyllocoptruta oleivora, Boophilus spp., Rhipicephalus spp., Amblyomma spp., Hyalomma spp., Ixodes spp., Psoroptes spp., Chorioptes spp., Sarcoptes spp., Tarsonemus spp., Bryobia praetiosa, Panonychus spp. and Tetranychus spp..

The active compounds according to the invention are distinguished by a powerful insecticidal and acaricidal activity.

5

15

20

They can be employed particularly successfully for combating plant-injurious insects, such as, for example, against the larvae of the mustard beetle (Phaedon cochleariae) or against the larvae of the green rice leafhopper (Nephotettix cincticeps) against the caterpillars of the diamond-back moth Plutella maculipennis.

- The active compounds according to the invention can be furthermore used as defoliants, agents for destroying broad-leaved plants and, especially, as weed-killers. By weeds, in the broadest sense, there are to be understood all plants which grow in locations where they are undesired. Whether the substances according to the invention act as total or selective herbicides depends essentially on the amount used.
- The active compounds according to the invention can be used, for example, in connection with the following plants:

Dicotyledon weeds of the genera: Sinapis, Lepidium, Galium, Stellaria, Matricaria,
 Anthemis, Galinsoga, Chenopodium, Urtica, Senecio, Amaranthus, Portulaca,
 Xanthium, Convolvulus, Ipomoea, Polygonum, Sesbania, Ambrosia, Cirsium, Carduus,
 Sonchus, Solanum, Rorippa, Rotala, Lindernia, Lamium, Veronica, Abutilon, Emex,
 Datura, Viola, Galeopsis, Papaver and Centaurea.

<u>Dicotyledon cultures of the genera:</u> Gossypium, Glycine, Beta, Daucus, Phaseolus, Pisum, Solanum, Linum, Ipomoea, Vicia, Nicotiana, Lycopersicon, Arachis, Brassica, Lactuca, Cucumis and Cucurbita.

- Monocotyledon weeds of the genera: Echinochloa, Setaria, Panicum, Digitaria, Phleum, Poa, Festuca, Eleusine, Brachiaria, Lolium, Bromus, Avena, Cyperus, Sorghum, Agropyron, Cynodon, Monochoria, Fimbristylis, Sagittaria, Eleocharis, Scirpus, Paspalum, Ischaemum, Sphenoclea, Dactyloctenium, Agrostis, Alopecurus and Apera.
- 25 Monocotyledon cultures of the genera: Oryza, Zea, Triticum, Hordeum, Avena, Secale, Sorghum, Panicum, Saccharum, Ananas, Asparagus and Allium.

However, the use of the active compounds according to the invention is in no way restricted to these genera, but also extends in the same manner to other plants.

The compounds are suitable, depending on the concentration, for the total combating of weeds, for example on industrial terrain and rail tracks, and on paths and squares with or without tree plantings. Equally, the compounds can be employed for combating weeds in perennial cultures, for example afforestations, decorative tree plantings, orchards, vineyards, citrus groves, nut orchards, banana plantations, coffee plantations, tea plantations, rubber plantations, oil palm plantations, cocoa plantations, soft fruit plantings and hopfields, and for the selective combating of weeds in annual cultures.

The active compounds according to the invention can be employed particularly successfully for combatting monocotyledon and dicotyledon weeds in monocotyledon and dicotyledon cultures, such as, for example, wheat, maize or soya beans.

The active compounds can be converted into the customary formulations, such as solutions, emulsions, suspensions, powders, foams, pastes, granules, aerosols, natural and synthetic materials impregnated with active compound, and very fine capsules in polymeric substances.

These formulations are produced in a known manner, for example by mixing the active compounds with extenders, that is liquid solvents, liquified gases under pressure and/or solid carriers, optionally with the use of surface-active agents, that is emulsifying agents and/or dispersing agents and/or foam-forming agents.

In the case of the use of water as an extender, organic solvents can, for example, also be used as auxiliary solvents. As liquid solvents, there are suitable in the main: aromatics, such as xylene, toluene or alkylnaphthalenes, chlorinated aromatics and chlorinated aliphatic hydrocarbons, such as chlorobenzenes, chloroethylenes or methylene chloride, aliphatic hydrocarbons, such as cyclohexane or paraffins, for example petroleum fractions, mineral and vegetable oils, alcohols, such as butanol or glycol as well as their ethers and esters, ketones, such as acetone, methyl ethyl ketone, methyl isobutyl ketone or cyclohexanone, strongly polar solvents, such as

5

15

20

25

÷

dimethylformamide and dimethyl sulphoxide, as well as water.

As solid carriers there are suitable: for example ammonium salts and ground natural minerals, such as kaolins, clays, talc, chalk, quartz, attapulgite, montmorillonite or diatomaceous earth, and ground synthetic minerals, such as highly disperse silica, alumina and silicates; as solid carriers for granules there are suitable: for example crushed and fractionated natural rocks such as calcite, marble, pumice, sepiolite and dolomite, as well as synthetic granules of inorganic and organic meals, and granules of organic material such as sawdust, coconut shells, maize cobs and tobacco stalks; as emulsifying and/or foam forming agents there are suitable: for example non-ionic and anionic emulsifiers, such as polyoxyethylene fatty acid esters, polyoxyethylene fatty alcohol ethers, for example alkylaryl polyglycol ethers, alkylsulphonates, alkyl sulphates, arylsulphonates as well as albumen hydrolysis products; as dispersing agents there are suitable: for example lignin-sulphite waste liquors and methylcellulose.

Adhesives such as carboxymethylcellulose and natural and synthetic polymers in the form of powders, granules or latexes, such as gum arabic, polyvinyl alcohol and polyvinyl acetate, as well as natural phospholipids, such as cephalins and lecithins, and synthetic phospholipids, can be used in the formulations. Further additives can be mineral and vegetable oils.

It is possible to use colorants such as inorganic pigments, for example iron oxide, titanium oxide and Prussian Blue, and organic dyestuffs, such as alizarin dyestuffs, azo dyestuffs and metal phthalocyanine dyestuffs, and trace nutrients such as salts of iron, manganese, boron, copper, cobalt, molybdenum and zinc.

The formulations in general contain between 0.1 and 95 per cent by weight of active compound, preferably between 0.5 and 90%.

25 For combating weeds, the active compounds according to the invention, as such or in the form of their formulations, can also be used as mixtures with known herbicides, finished formulations or tank mixes being possible.

5

10

15

Suitable herbicides for the mixtures are known herbicides, for example anilides such as. for example, diflufenican and propanil; arylcarboxylic acids such as, for example, dichloropicolinic acid, dicamba and picloram; aryloxyalkanoic acids such as, for example, 2,4-D, 2,4-DB, 2,4-DP, fluroxypyr, MCPA, MCPP and triclopyr, aryloxyphenoxy-alkanoic esters such as, for example, diclofop-methyl, fenoxaprop-ethyl, fluazifop-butyl, haloxyfop-methyl and quizalofop-ethyl; azinones such as, for example, chloridazon and norflurazon; carbamates such as, for example, chlorpropham, desmedipham, phenmedipham and propham; chloroacetanilides such as, for example, alachlor, acetochlor, butachlor, metazachlor, metolachlor, pretilachlor and propachlor; dinitroanilines such as, for example, oryzalin, pendimethalin and trifluralin; diphenyl ethers such as, for example, acifluorfen, bifenox, fluoroglycofen, fomesafen, halosafen, lactofen and oxyfluorfen; ureas such as, for example, chlortoluron, diuron, fluometuron, isoproturon, linuron and methabenzthiazuron; hydroxylamines such as, for example, alloxydim, clethodim, cycloxydim, sethoxydim and tralkoxydim; imidazolinones such as, for example, imazethapyr, imazamethabenz, imazapyr and imazaquin; nitriles such as, for example, bromoxynil, dichlobenil and ioxynil; oxyacetamides such as, for example, mefenacet; sulphonylureas such as, for example, amidosulfuron, bensulfuronchlorimuron-ethyl, methyl, chlorsulfuron, cinosulfuron, metsulfuron-methyl. nicosulfuron, primisulfuron, pyrazosulfuron-ethyl, thifensulfuron-methyl, triasulfuron and tribenuron-methyl; thiocarbamates such as, for example, butylate, cycloate, diallate, EPTC, esprocarb, molinate, prosulfocarb, thiobencarb and tri-allate; triazines such as, for example, atrazine, cyanazine, simazine, simetryn, terbutryn and terbutylazine; triazinones such as, for example, hexazinone, metamitron and metribuzin; others such as, for example, aminotriazole, benfuresate, bentazone, cinmethylin, clomazone, clopyralid, difenzoquat, dithiopyr, ethofumesate, fluorochloridone, glufosinate, glyphosate, isoxaben, pyridate, quinchlorac, quinmerac, sulphosate and tridiphane.

Mixtures with other known active compounds, such as fungicides, insecticides, acaricides, nematicides, bird repellants, plant nutrients and agents which improve soil structure, are also possible.

The active compounds can be used as such, in the form of their formulations or in the

5

10

15

20

25

use forms prepared therefrom by further dilution, such as ready-to-use solutions, suspensions, emulsions, powders, pastes and granules. They are used in the customary manner, for example by watering, spraying, atomizing or scattering.

The active compounds according to the invention can be applied either before or after emergence of the plants.

They can also be incorporated into the soil before sowing.

5

10

15

20

25

The amount of active compound used can vary within a substantial range. It depends essentially on the nature of the desired effect. In general, the amounts used are between 0.001 and 10 kg of active compound per hectare of soil surface, preferably between 0.005 and 5 kg per ha.

When the active compounds according to the invention are used as insecticides, they can, again, be present in their commercially available formulations and in the use forms, prepared from these formulations, as a mixture with other active compounds, such as insecticides, attractants, sterilizing agents, acaricides, nematicides, fungicides, growth-regulating substances or herbicides. The insecticides include, for example, phosphates, carbamates, carboxylates, chlorinated hydrocarbons, phenylureas and substances produced by microorganisms.

When the active compounds according to the invention are used as insecticides, they can furthermore be present in their commercially available formulations and in the use forms, prepared from these formulations, as a mixture with synergistic agents. Synergistic agents are compounds which increase the action of the active compounds, without it being necessary for the synergistic agent added to be active itself.

The active compound content of the use forms prepared from the commercially available formulations can vary within wide limits. The active compound concentration of the use forms can be from 0.0000001 to 95 per cent by weight of active compound, preferably between 0.0001 and 1 per cent by weight.

# 2158996

The compounds are employed as insecticides in a customary manner appropriate for the use forms.

Preparation and use of the active compounds according to the invention can be seen from the examples which follow.

#### Preparation examples

## Example 1

5

10

Br 
$$CF_3$$
 +  $P_1$   $P_2$   $P_3$   $P_4$   $P_5$   $P_6$   $P_6$   $P_7$   $P_8$   $P_8$ 

2.66 g (0.01 mol) 2-trifluoromethyl-bromo-pyridino-[1H]-imidazole (A1/A2) and 1.75 ml (0.0125 mol) of triethylamine are dissolved in 100 ml of dichloromethane. 1.25 ml (0.0125 mol of chloromethyl methyl ether are added dropwise to this solution, the mixture is subsequently heated at reflux temperature, and stirring is continued for 16 hours at reflux temperature. For working up, the cooled reaction mixture is washed three times using 30 ml of water in each case, dried over MgSO<sub>4</sub> and concentrated in vacuo and the residue is purified by chromatography on silica gel (eluent: dichloromethane).

2.40 g (74 % of theory) of 1-ethoxymethyl-2-trifluoromethyl-bromopyridino imidazole are obtained as a regio isomer mixture (B1/B2) in a ratio of 60:40 (m.p.: 68°C).

<sup>1</sup>H NMR (CDCl<sub>3</sub>/tetramethylsilane):  $\delta$  = 5.68 (s, 2H); 5.85 (s, 2H) ppm (in each case N-CH<sub>2</sub>-O-).

15 The isomers can be separated by recrystallization with an ether/petroleum ether mixture.

The compounds listed in the table which follows are obtained analogously.

Table II

$$A$$

$$A$$

$$A$$

$$A$$

$$A$$

$$A$$

$$A$$

$$CH-R$$

$$A$$

$$R$$

Example No.	A'	A <sup>2</sup>	A³	A <sup>4</sup>	R'	R²	R³	Physical data
2 .	N	СН	СН	СН	Н	OC₂H₅	CF,	m.p.: 92°C
3	СН	СН	CH	N	Н	OC2H3	CF <sub>3</sub>	m.p.: 170°C
4	СН	N	СН	СН	Н	OC <sub>2</sub> H <sub>5</sub>	CF <sub>3</sub>	<sup>1</sup> H NMR*: 5.89 (s, 2H)
5	СН	СН	N	СН	Н	OC₂H,	CF <sub>3</sub>	'H NMR*: 6.08 (s, 2H)
6	N	СН	CBr	СН	Н	CH,	CF <sub>3</sub>	'H NMR': 5.90 (s, 2H)
7	СН	CBr	СН	N	Н	COOE:	CF <sub>3</sub>	'H NMR*: 6.08 (s, 2H)
8	N	СН	CBr	СН	Н	N COOE:	CF <sub>3</sub>	'H NMR*: 5.90 (s, 2H)
9	СН	СВг	СН	N	Н	N COOE:	CF <sub>3</sub>	<sup>1</sup> H NMR*: 6.10 (s, 2H)

Table II (continued)

Example No.	A <sup>1</sup>	A <sup>2</sup>	A³	A <sup>4</sup>	R¹	R²	R³	Physical data
						-		
10	N	СН	CBr	СН	Н	n-Propyl	CF <sub>3</sub>	<sup>1</sup> H NMR*: 5.89 (s, 2H)
11	СН	CBr	СН	N	Н	N COOE:	CF,	<sup>1</sup> H NMR*: 6.09 (s, 2H)
12	N	СН	СВг	СН	Н	N COOR	CF <sub>3</sub>	¹H NMR*: 5.90 (s, 2H)
13	СН	СВг	СН	N	Н	N COOR	CF <sub>3</sub>	'H NMR*: 6.11 (s, 2H)
14	N	СН	СН	СН	Н	OC₂H,	CHF <sub>2</sub>	<sup>1</sup> H NMR*: 5.91 (s, 2H)
15	СН	СН	СН	N	Н	OC <sub>2</sub> H,	CHF <sub>2</sub>	'H NMR*: 6.14 (s, 2H)
16	СН	N	СН	СН	Н	Et COOEt	CF <sub>3</sub>	¹H NMR*: 5.83 (s, 2H) m.p.: 120°C
17	СН	СН	N	СН	Н	N COOE:	CF,	'H NMR': 6.03 (s, 2H)
18	N	СН	СН	СН	Н	CH=CH₂	CF,	<sup>1</sup> H NMR*: 5.35 (d, J = 3Hz, 2H)

Table II (continued)

Example No.	A¹	A <sup>2</sup>	A³	A <sup>4</sup>	R¹	R²	R³	Physical data
19	N	СН	СН	СН	Н	COCH <sub>3</sub>	CF <sub>3</sub>	m.p.: 148- 150°C
20	N	СН	СН	СН	Н	CN	CHF₂	'H NMR*: 5.48 (s, 2H)
21	N	СН	СН	СН	Н	N COOE:	CF <sub>3</sub>	'H NMR*: 5.89 (s, 2H)
22	СН	СН	СН	N	Н	N COOE:	CF <sub>3</sub>	<sup>1</sup> H NMR*: 6.12 (s, 2H)
23	N	СН	СН	СН	н	Me N COORS	CF <sub>3</sub>	'H NMR*: 5.94 (s, 2H)
24	СН	СН	СН	N	Н	Me COORI	CF <sub>3</sub>	'H NMR*: 6.12 (s, 2H)
25	N	СН	СН	СН	Н	N COOR	CHF₂	¹H NMR˚: 5.88 (s, 2H) m.p.: 119°C
26	СН	СН	СН	N	Н	N COOOR	CHF <sub>2</sub>	'H NMR*: 6.02 (s, 2H)
27	N	СН	СН	СН	Н	COOP	CHF <sub>2</sub>	'H NMR*: 5.93 (s, 2H)

Table II (continued)

Example No.	A <sup>1</sup>	A <sup>2</sup>	A³	A <sup>4</sup>	R¹	R²	R³	Physical data
						:		
28	СН	СН	СН	N	Н	N EI	CHF <sub>2</sub>	<sup>1</sup> H NMR*: 6.14 (s, 2H) m.p.: <b>84</b> °C
29	N	CCI	СН	СН	H	N Et	CF,	<sup>1</sup> H NMR*: 5.88 (s, 2H)
30	СН	СН	CCI	N	Н	N EI	CF <sub>3</sub>	<sup>1</sup> H NMR*: 6.02 (s, 2H)
31	N CHR₁ R₂	СН	СН	СН	Н	СИ	CF <sub>3</sub>	m.p.: 184- 186°C

 $<sup>^{\</sup>bullet}$  <sup>1</sup>H NMR spectra were recorded in deuterochloroform (CDCl<sub>3</sub>) with tetramethylsilane (TMS) as the internal standard. The data given is the chemical shift as  $\delta$ -value in ppm; in all cases, the N-CH<sub>2</sub>R<sup>1</sup>R<sup>2</sup> proton shift is given.

## Example A:

## Pre-emergence test

Solvent:

5 parts by weight of acetone

Emulsifier:

1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, one part by weight of active compound is mixed with the stated amount of solvent, the stated amount of emulsifier is added and the concentrate is diluted with water to the desired concentration.

Seeds of the test plants are sown in normal soil and, after 24 hours, watered with the preparation of the active compound. It is expedient to keep constant the amount of water per unit area. The concentration of the active compound in the preparation is of no importance, only the amount of active compound applied per unit area being decisive. After three weeks, the degree of damage to the plants is rated in % damage in comparison to the development of the untreated control. The figures denote:

0% = no action (like untreated control) 100% = total destruction

15

20

10

In this test, a clearly superior activity combined with a similarly good crop plant selectivity is shown by the compounds of Preparation Examples (1) and (6), for example in wheat crops at application rates of 1,000 g per hectare when applied against weeds such as Chenopodium (95-100 %), Galinsoga (95-100 %), Matricaria (90-95 %), Portulaca (100 %), Stellaria (100 %) and Viola (90-95 %), the wheat remaining unharmed (0 %).

Table III

Pre-emergence test/greenhouse

Active comp.	Applica- tion rate in g/ha	Wheat	Cheno- podium	Galins- oga	Matri c-aria	Portu- laca	Stell -aria	Viola
afoch(	1000	0	100	100	95	100	100	95
BC TOCK	1000	0	95	95	90	100	100	90

#### Example B:

#### Post-emergence test

Solvent:

10

5 parts by weight of acetone

Emulsifier:

1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compound, one part by weight of active compound is mixed with the stated amount of solvent, the stated amount of emulsifier is added and the concentrate is diluted with water to the desired concentration.

Test plants which have a height of 5 - 15 cm are sprayed with the preparation of the active compound in such a way as to apply the particular amounts of active compound desired per unit area. After three weeks, the degree of damage to the plants is rated in % damage in comparison to the development of the untreated control. The figures denote:

0% = no action (like untreated control) 100% = total destruction

In this test, a clearly superior activity and crop plant selectivity is shown by the compounds of Preparation Examples (1), (6) and (12), for example in wheat crops at application rates of 250 g per hectare when used against weeds such as Datura (90-100 %), Helianthus (90-100 %), Portulaca (90-100 %), Sinapis (100 %) and Solanum (80-100 %), the wheat remaining unharmed (0 %).

Table IV

Post-emergence test/greenhouse

Active compound	Appli- tion rate in g/ha	Wheat	Datura	Helian- thus	Portu- laca	Sinapis	Solan- um
GLOCK (I)	250	0	90	90	90	- 100	80
- Pack	250	0	100	100	100	100	100
BC TOCAL	250	0	100	100	100	100	100

## Example C:

#### Phaedon larvae test

Solvent:

10

7 parts by weight of dimethylformamide

Emulsifier:

1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compond, one part by weight of active compound is mixed with the stated amount of solvent and the stated amount of emulsifier, and the concentrate is diluted with water to the desired concentration.

Cabbage leaves (Brassica oleracea) are treated by being dipped into the preparation of active compound of the desired concentration and are infested with mustard beetle larvae (Phaedon cochleariae) while the leaves are still moist.

After the specified periods of time, the destruction in % is determined. 100% means that all the beetle larvae have been killed; 0% means that none of the beetle larvae have been killed.

In this test, for example the compound of Preparation Example (1) shows a degree of destruction of 100 % after 7 days at an active compound concentration 0.1 %.

100

Table V

Phaedon larvae test (plant-injurious insects)

Active compounds

Active Degree of

compound destruction in %

concentration after 7<sup>d</sup>

in %

0.1

$$H_3C^ONN$$
 $H_3C^N$ 
 $H_3C^N$ 

## Example D:

#### Plutella test

Solvent:

10

15

5 parts by weight of dimethylformamide

Emulsifier:

1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compond, one part by weight of active compound is mixed with the stated amount of solvent and the stated amount of emulsifier, and the concentrate is diluted with water to the desired concentration.

Cabbage leaves (Brassica oleracea) are treated by being dipped into the preparation of active compound of the desired concentration and are infested with caterpillars of the diamond-back moth (Plutella maculipennis) while the leaves are still moist.

After the specified periods of time, the destruction in % is determined. 100% means that all the caterpillars have been killed; 0% means that none of the caterpillars have been killed.

In this test, for example the compound of Preparation Example (1) shows a degree of destruction of 100 % after 7 days at an active compound concentration of 0.1 %.

Table VI

Plutella test (plant-injurious insects)

Active compounds

Active compound Degree of

destruction in %

concentration

after 7d

in %

0.1

100

(1)

#### Example E:

#### Nephotettix test

Solvent:

10

7 parts by weight of dimethylformamide

Emulsifier:

1 part by weight of alkylaryl polyglycol ether

To produce a suitable preparation of active compond, one part by weight of active compound is mixed with the stated amount of solvent and the stated amount of emulsifier, and the concentrate is diluted with water to the desired concentration.

Rice seedlings (Oryza sativa) are treated by being dipped into the preparation of active compound of the desired concentration and are infested with larvae of the green rice leafhopper (Nephotettix cincticeps) while the leaves are still moist.

After the specified periods of time, the destruction in % is determined. 100% means that all the leafhoppers have been killed; 0% means that none of the leafhoppers have been killed.

In this test, for example the following compounds of preparation examples (1) and (10) show degrees of destruction of up to 100 % after 6 days at an active compound concentration of 0.1 %.

5

## Nephotettix test

# (plant-injurious insects)

Active	Degree of
compound	destruction in %
concentration	after 6d
in %	
0.1	100
	concentration in %

#### Patent Claims

1. New substituted hetero-fused imidazoles of the general formula (1)

in which

R<sup>1</sup> represents hydrogen or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl, alkoxy and aryl,

R<sup>2</sup> represents hydroxyl, cyano or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, amino, aminocarbonyl, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy, dialkoxyphosphonyl, (hetero)aryl, (hetero)arylcarbonyl, (hetero)aryloxycarbonyl, (hetero)arylcarbonyloxy and (hetero)arylaminocarbonyloxy,

- R³ represents cyano, halogen or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl, alkenyl, alkinyl, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy, alkenyloxy, alkoxy, alkinyloxy, dialkoxyphosphonyl, amino, aminocarbonyl and aryl,
- A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> and A<sup>4</sup> in each case represent N(nitrogen), N-CHR<sup>1</sup>R<sup>2</sup> or CX, the hetero-fused ring having at least one, but not more than two, nitrogen atoms simultaneously and all positional isomers being possible, so that

20

15

5

CX1, CX2, CX3 exist in the case of one nitrogen atom and

CX<sup>1</sup> and CX<sup>2</sup> exist in the case of two nitrogen atoms, and, when either A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> or A<sup>4</sup> represent N-CHR<sup>1</sup>R<sup>2</sup>, the imidazole ring exists only in monosubstituted form (R<sup>3</sup>),

#### 5 where

10

15

20

- X1, X2 and X3 in each case independently of one another represent hydrogen, halogen, cyano, nitro or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl, alkoxy, alkylthio, alkylsulphinyl, alkylsulphonyl and cycloalkyl, or represents hydroxycarbonyl, alkylcarbonyl, alkoxycarbonyl, cycloalkyloxycarbonyl, or represents in each case optionally substituted amino or aminocarbonyl, or represents in each case optionally substituted aryl, aryloxy, arylthio, arylsulphinyl, arylsulphonyl, arylsulphonyloxy. arylcarbonyl, aryloxycarbonyl, arylazo or arylthiomethylsulphonyl, but where at least one of the substitutents X1, X2 or X3 represents halogenoalkyl, halogenoalkylylinio, halogenoalkylsulphinyl, halogenoalkylsulphonyl, alkylsulphonyl, or represents optionally substituted fused dioxyalkylene, or represents hydroxycarbonyl, alkylcarbonyl, alkoxycarbonyl, cycloalkyloxycarbonyl, or represents in each case optionally substituted amino or aminocarbonyl, or represents in each case optionally substituted aryl, arylthio, arylsulphinyl, arylsulphonyl, arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylazo arylthiomethylsulphonyl.
- New substituted hetero-fused imidazoles of the general formula (I) according to Claim 1, characterized in that
  - R¹ represents hydrogen or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl and alkoxy, each of which has 1 to 8 carbon atoms, or represents phenyl which

is optionally monosubstituted or polysubstituted by identical or different substituents, suitable substituents being:

5

10

15

20

25

halogen, cyano, nitro in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of which has 1 to 6 carbon atoms, in each case straight-chain or branched halogenoalkyl. halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl halogenoalkylsulphonyl each of which has 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, in each case straight-chain or branched alkoxyalkyl, alkoxyalkoxy, alkanoyl, alkoxycarbonyl or alkoximinoalkyl each of which has 1 to 6 carbon atoms in the individual alkyl moieties, or divalent dioxyalkylene having 1 to 5 carbon atoms which is optionally monosubstituted or polysubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 6 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, or phenyl which is optionally monosubstituted or polysubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 6 carbon atoms and straightchain or branched halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms,

R<sup>2</sup> represents hydroxyl, cyano or a straight-chain or branched radical from the series consisting of alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy or dialkoxyphosphonyl, each of which has up to 8 carbon atoms in the individual alkyl or alkenyl or alkinyl moieties and each of these radicals optionally being monosubstituted or polysubstituted by identical or different substituents, suitable substituents in each case being:

fluorine, chlorine, bromine, iodine, straight-chain or branched alkoxy having 1 to 8 carbon atoms, or aryl having 6 to 10 carbon atoms or heteroaryl having 2 to 9 carbon atoms and 1 to 5 hetero atoms (in

particular nitrogen, oxygen and/or sulphur), these aryl or heteroaryl substituents in each case optionally being monosubstituted or polysubstituted by identical or different substituents and suitable aryl or heteroaryl substituents being those mentioned in the case of R<sup>1</sup>.

R<sup>2</sup> furthermore represents amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

> formyl, straight-chain or branched alkyl having 1 to 8 carbon atoms, straight-chain or branched alkenyl having 2 to 8 carbon atoms, straightchain or branched alkylsulphonyl having 1 to 8 carbon atoms, carbamoyl, thiocarbamoyl or sulphamoyl, each of which is monosubstituted or disubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 8 carbon atoms, cycloalkyl, cycloalkylcarbonyl or cycloalkyloxycarbonyl, each of which has 3 to 8 carbon atoms in the cycloalkyl moiety, alkylcarbonyl, alkenylcarbonyl, alkoxycarbonyl, alkenyloxycarbonyl, alkylthio-carbonyl, alkoxy-thiocarbonyl or alkylthiothiocarbonyl, each of which has 1 to 8 carbon atoms in the individual straight-chain or branched alkyl moieties, in each case divalent and cyclized alkanediylcarbonyl or alkanediyloxycarbonyl, each of which has 2 to 6 carbon atoms in the alkanediyl moiety, arylalkyl, arylalkylcarbonyl or arylalkyloxycarbonyl, each of which has 6 to 10 carbon atoms in the aryl moiety and 1 to 8 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted in the aryl moiety by identical or different substituents, or aryl, arylcarbonyl or aryloxycarbonyl, each of which has 6 to 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted or polysubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of  $R^{1}$ ,

30 R<sup>2</sup> furthermore represents aryl, arylcarbonyl, arylcarbonyl, arylcarbonyloxy

Le A 29 565

5

10

15

20

or arylaminocarbonylaminocarbonyloxy, each of which has 6 to 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted or polysubstituted by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>,

R<sup>2</sup> furthermore represents heteroaryl, heteroarylcarbonyl, heteroarylcarbonyl, heteroarylcarbonyloxy or heteroarylaminocarbonylaminocarbonyloxy, each of which has 2 to 9 carbon atoms and 1 to 5 identical or different hetero atoms (in particular nitrogen, oxygen and/or sulphur) in the heteroaryl moiety and each of which is optionally monosubstituted or polysubstituted by identical or different substituents, suitable heteroaryl substituents in each case being the aryl substituents

mentioned in the case of R<sup>1</sup>,

represents cyano, fluorine, chlorine, bromine, iodine or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of cycloalkyl, alkyl, alkenyl, alkinyl, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy, alkenyloxy, alkoxy, alkenyloxy, each of which has up to 8 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties, suitable substituents in each case being: fluorine, chlorine, bromine, iodine, straight-chain or branched alkoxy having 1 to 8 carbon atoms, or aryl having 6 to 10 carbon atoms or heteroaryl having 2 to 9 carbon atoms and 1 to 5 hetero atoms (in particular nitrogen, oxygen and/or sulphur), each of these aryl or heteroaryl radicals optionally being monosubstituted or polysubstituted by identical or different substituents and suitable aryl or heteroaryl substituents being those mentioned in the case of R<sup>1</sup>,

25

15

20

R³ furthermore represents amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

formyl, straight-chain or branched alkyl having 1 to 8 carbon atoms,

straight-chain or branched alkenyl having 2 to 8 carbon atoms, straightchain or branched alkylsulphonyl having 1 to 8 carbon atoms, carbamovl. thiocarbamoyl or sulphamoyl, each of which is monosubstituted or disubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 8 carbon atoms, cycloalkyl, cycloalkylcarbonyl or cycloalkyloxycarbonyl, each of which has 3 to 8 carbon atoms in the cycloalkyl moiety, alkylcarbonyl, alkenylcarbonyl, alkoxycarbonyl, alkenyloxycarbonyl, alkylthio-carbonyl, alkoxy-thiocarbonyl or alkylthiothiocarbonyl, each of which has 1 to 8 carbon atoms in the individual straight-chain or branched alkyl moieties, in each case divalent and cyclized alkanediylcarbonyl or alkanediyloxycarbonyl, each of which has 2 to 6 carbon atoms in the alkanediyl moiety, arylalkyl, aryalkylcarbonyl or arylalkyloxycarbonyl, each of which has 6 to 10 carbon atoms in the aryl moiety and 1 to 8 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or polysubstituted in the aryl moiety by identical or different substituents, or aryl, arylcarbonyl or aryloxycarbonyl, each of which has 6 to 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted or polysubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of  $R^{1}$ 

R³ furthermore represents aryl having in each case 6 to 10 carbon atoms in the aryl moiety which is in each case optionally monosubstituted or polysubstituted by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R¹,

A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> and A<sup>4</sup> in each case represent N(nitrogen), N-CHR<sup>1</sup>R<sup>2</sup> or CX, the hetero-fused ring having at least one, but not more than two, nitrogen atoms simultaneously and all positional isomers being possible, so that

CX1, CX2, CX3 exist in the case of one nitrogen atom and

5

10

15

20

CX<sup>1</sup> and CX<sup>2</sup> exist in the case of two nitrogen atoms, and, when either A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> or A<sup>4</sup> represent N-CHR<sup>1</sup>R<sup>2</sup>, the imidazole ring exists only in monosubstituted form (R<sup>3</sup>), and

X1, X2 and X3 in each case independently of one another represent hydrogen, fluorine, chlorine, bromine, iodine, cyano, nitro, in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of which has 1 to 8 carbon atoms, cycloalkyl having 3 to 8 carbon atoms, in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl, halogenoalkylsulphonyl, each of which has 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, or divalent dioxyalkylene having 1 to 5 carbon atoms which is optionally monosubstituted or polysubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, furthermore represent hydroxycarbonyl, in each case straight-chain or branched alkylcarbonyl or alkoxycarbonyl, each of which has 1 to 6 carbon atoms in the alkyl moiety, cycloalkyloxycarbonyl having 3 to 8 carbon atoms in the cycloalkyl moiety, or amino or aminocarbonyl, each of which is optionally monosubstituted or polysubstituted by identical or different substituents, suitable amino substituents in each case being:

in each case straight-chain or branched alkyl having 1 to 6 carbon atoms, halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 halogen atoms, alkoxyalkyl or alkylcarbonyl, each of which has 1 to 6 carbon atoms in the individual alkyl moieties, or arylcarbonyl, arylsulphonyl, arylaminocarbonyl or arylmethylsulphonyl, each of which has 6 to 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted or polysubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>;

X1, X2 and X3 furthermore represent aryl, aryloxy, arylthio, arylsulphinyl,

30

25

5

10

15

20

Le A 29 565

arylsulphonyl, arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylthiomethylsulphonyl or arylazo, each of which has 6 to 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted or polysubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>, and

10

5

15

20

25

30

where at least one of the substituents  $X^1$ ,  $X^2$  or  $X^3$  represents in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogeno-alkylsulphinyl, halogenoalkylsulphonyl, each of which has 1 to 6 carbon atoms and 1 to 13 identical or different halogen atoms, or represents straight-chain or branched alkylsulphonyl having 1 to 6 carbon atoms, or divalent dioxyalkylene having 1 to 5 carbon atoms which is optionally monosubstituted or polysubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, furthermore represents hydroxycarbonyl, in each case straight-chain or branched alkylcarbonyl or alkoxycarbonyl, each of which has 1 to 6 carbon atoms in the alkyl moiety, cycloalkyloxycarbonyl having 3 to 8 carbon atoms in the cycloalkyl moiety, or amino or aminocarbonyl, each of which is optionally monosubstituted or polysubstituted by identical or different substituents, suitable amino substituents in each case being:

in each case straight-chain or branched alkyl having 1 to 6 carbon atoms, halogenoalkyl having 1 to 6 carbon atoms and 1 to 13 halogen atoms, alkoxyalkyl or alkylcarbonyl, each of which has 1 to 6 carbon atoms in the individual alkyl moieties, or arylcarbonyl, arylsulphonyl, arylaminocarbonyl or arylmethylsulphonyl, each of which has 6 to 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted or polysubstituted by identical or different substituents in the aryl moiety, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>,

X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> furthermore represent aryl, arylthio, arylsulphinyl, arylsulphonyl, arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylthiomethylsulphonyl or arylazo, each of which has 6 to 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted or polysubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>.

 New substituted hetero-fused imidazoles of the general formula (I) according to Claim 1, characterized in that

R¹ represents hydrogen, or a straight-chain or branched radical from the series consisting of alkyl and alkoxy, each of which has 1 to 6 carbon atoms and each of which is unsubstituted or substituted, or represents phenyl which is optionally monosubstituted to trisubstituted by identical or different substituents, suitable substituents being:

fluorine, chlorine, bromine, iodine, cyano, nitro, in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of which has 1 to 4 carbon atoms, in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl or halogenoalkylsulphonyl, each of which has 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, in each case straight-chain or branched alkoxyalkyl, alkoxyalkoxy, alkanoyl, alkoxycarbonyl or alkoximinoalkyl, each of which has 1 to 4 carbon atoms in the individual alkyl moieties, divalent dioxyalkylene having 1 to 4 carbon atoms which is optionally monosubstituted to hexasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, or phenyl which is optionally monosubstituted to pentasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon

30

5

10

15

20

atoms and 1 to 9 identical or different halogen atoms, halogen in each case representing fluorine, chlorine, bromine or iodine,

R<sup>2</sup> represents hydroxyl, cyano, or a straight-chain or branched radical from the series consisting of alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy dialkoxyphosphonyl, each of which has up to 6 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties and each of which is optionally monosubstituted to pentasubstituted by identical or different substituents from the series consisting of fluorine, chlorine, bromine and iodine, or represents alkyl, alkenyl or alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylcarbonyl. alkoxycarbonyl, alkylcarbonyloxy dialkoxyphosphonyl, each of which has up to 6 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties and each of which is optionally monosubstituted to trisubstituted by identical or different substituents. suitable substituents in each case being:

straight-chain or branched alkoxy having 1 to 6 carbon atoms, or aryl having 6 or 10 carbon atoms or heteroaryl having 2 to 9 carbon atoms and 1 to 4 hetero atoms (in particular nitrogen, oxygen and/or sulphur), each of these aryl or heteroaryl radicals optionally being monosubstituted to trisubstituted by identical or different substituents and suitable aryl or heteroaryl substituents being those mentioned in the case of R<sup>1</sup>,

R<sup>2</sup> furthermore represents amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

formyl, straight-chain or branched alkyl having 1 to 6 carbon atoms, straight-chain or branched alkenyl having 2 to 6 carbon atoms, straight-chain or branched alkylsulphonyl having 1 to 6 carbon atoms, carbamoyl, thiocarbamoyl or sulphamoyl, each of which is optionally monosubstituted or disubstituted by identical or different straight-chain or branched alkyl

20

5

10

15

substituents having 1 to 6 carbon atoms, or cycloalkyl, cycloalkylcarbonyl or cycloalkyloxycarbonyl, each of which has 3 to 7 carbon atoms in the cycloalkyl moiety, alkylcarbonyl, alkenylcarbonyl, alkoxycarbonyl, alkenyloxycarbonyl, alkylthio-carbonyl, alkoxy-thiocarbonyl or alkylthiothiocarbonyl, each of which has 1 to 6 carbon atoms in the individual straight-chain or branched alkyl moieties, in each case divalent and cyclized alkanediylcarbonyl or alkanediyloxycarbonyl, each of which has 2 to 5 carbon atoms in the alkanediyl moiety, arylalkyl, arylalkylcarbonyl or arylalkyloxycarbonyl, each of which has 6 or 10 carbon atoms in the aryl moiety and 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted to trisubstituted in the aryl moiety by identical or different substituents, or aryl, arylcarbonyl or aryloxycarbonyl, each of which has 6 or 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted to trisubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R1,

5

10

15

20

25

R<sup>2</sup> furthermore represents aryl, arylcarbonyl, arylcarbonyl, arylcarbonyloxy or arylaminocarbonylaminocarbonyloxy, each of which has 6 or 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted to pentasubstituted by identical or different substituents, suitable aryl substituents in each case being those mentioned under R<sup>1</sup>,

R<sup>2</sup> furthermore represents heteroaryl, heteroarylcarbonyl, heteroaryloxycarbonyl, heteroarylcarbonyloxy or heteroarylamino-carbonylaminocarbonyloxy, each of which has 2 to 9 carbon atoms and 1 to 4 identical or different hetero atoms (in particular nitrogen, oxygen and/or sulphur) in the heteroaryl moiety and each of which is optionally monosubstituted to pentasubstituted by identical or different substituents, suitable heteroaryl substituents in each case being the aryl substituents mentioned in the case of R<sup>1</sup>.

R<sup>3</sup> represents cyano, fluorine, chlorine, bromine, iodine, or a straight-chain or
Le A 29 565

- 68 -

branched radical from the series consisting of alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl and alkylcarbonyloxy, each of which has up to 6 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties and which is optionally monosubstituted to pentasubstituted by identical or different substituents from the series consisting of fluorine, chlorine, bromine and iodine, or represents cycloalkyl, alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy or dialkoxy-phosphonyl, each of which has up to 6 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties and each of which is optionally monosubstituted to trisubstituted by identical or different substituents, suitable substituents in each case being:

fluorine, chlorine, bromine, iodine, straight-chain or branched alkoxy having 1 to 6 carbon atoms, or aryl having 6 or 10 carbon atoms or heteroaryl having 2 to 9 carbon atoms and 1 to 4 hetero atoms (in particular nitrogen, oxygen and/or sulphur), each of these aryl or heteroaryl radicals optionally being monosubstituted to trisubstituted by identical or different substituents, suitable aryl or heteroaryl substituents being those mentioned in the case of R<sup>1</sup>,

R<sup>3</sup> furthermore represents amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

formyl, straight-chain or branched alkyl having 1 to 6 carbon atoms, straight-chain or branched alkenyl having 2 to 6 carbon atoms, straight-chain or branched alkylsulphonyl having 1 to 6 carbon atoms, carbamoyl, thiocarbamoyl or sulphamoyl, each of which is optionally monosubstituted or disubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 6 carbon atoms, or cycloalkyl, cycloalkylcarbonyl, or cycloalkyloxycarbonyl, each of which has 3 to 7 carbon atoms in the cycloalkyl moiety, alkylcarbonyl, alkenylcarbonyl, alkoxycarbonyl,

30

25

5

10

15

alkenyloxycarbonyl, alkylthio-carbonyl, alkoxy-thiocarbonyl or alkylthio-thiocarbonyl, each of which has 1 to 6 carbon atoms in the individual straight-chain or branched alkyl moieties, in each case divalent and cyclized alkanediylcarbonyl or alkanediyloxycarbonyl, each of which has 2 to 5 carbon atoms in the alkanediyl moiety, or arylalkyl, arylalkylcarbonyl or arylalkyloxycarbonyl, each of which has 6 or 10 carbon atoms in the aryl moiety and 1 to 6 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted to trisubstituted by identical or different substituents in the aryl moiety, or aryl, arylcarbonyl or aryloxycarbonyl, each of which has 6 or 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted to trisubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>.

- R<sup>3</sup> furthermore represents aryl having in each case 6 or 10 carbon atoms in the aryl moiety which is in each case optionally monosubstituted to pentasubstituted by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>,
- A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> and A<sup>4</sup> in each case represent N(nitrogen), N-CHR<sup>1</sup>R<sup>2</sup> or CX, the hetero-fused ring having at least one, but not more than two, nitrogen atoms simultaneously and all positional isomers being possible, so that
  - CX1, CX2, CX3 exist in the case of one nitrogen atom and
  - CX<sup>1</sup> and CX<sup>2</sup> exist in the case of two nitrogen atoms, and, when either A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> or A<sup>4</sup> represent N-CHR<sup>1</sup>R<sup>2</sup>, the imidazole ring exists only in monosubstituted form (R<sup>3</sup>), and
  - X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> in each case independently of one another represent hydrogen, fluorine, chlorine, bromine, cyano, nitro, in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of

5

10

15

20

which has 1 to 6 carbon atoms, cycloalkyl having 3 to 7 carbon atoms, in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkyl-sulphinyl, halogenoalkylsulphonyl, each of which has 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, or divalent dioxyalkylene having 1 to 4 carbon atoms which is optionally monosubstituted to hexasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, furthermore represent hydroxycarbonyl, in each case straight-chain or branched alkylcarbonyl or alkoxycarbonyl, each of which has 1 to 4 carbon atoms in the alkyl moiety, cycloalkyloxycarbonyl having 3 to 7 carbon atoms in the cycloalkyl moiety, or amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable amino substituents in each case being:

in each case straight-chain or branched alkyl having 1 to 4 carbon atoms, halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 halogen atoms, alkoxyalkyl or alkylcarbonyl, each of which has 1 to 4 carbon atoms in the individual alkyl moieties, or arylcarbonyl, arylsulphonyl, arylaminocarbonyl or arylmethylsulphonyl, each of which has 6 or 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted to pentasubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>;

X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> furthermore represent aryl, aryloxy, arylthio, arylsulphinyl, arylsulphonyl, arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylthiomethylsulphonyl or arylazo, each of which has 6 or 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted to pentasubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>, and

25

5

10

15

20

where at least one of the substituents  $X^1$ ,  $X^2$  and  $X^3$  represents in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl, halogenoalkylsulphonyl, each of which has 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, straight-chain or branched alkylsulphonyl having 1 to 4 carbon atoms, or divalent dioxyalkylene having 1 to 4 carbon atoms which is optionally monosubstituted to hexasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 4 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 identical or different halogen atoms, furthermore represents hydroxycarbonyl, in each case straight-chain or branched alkylcarbonyl or alkoxycarbonyl, each of which has 1 to 4 carbon atoms in the alkyl moiety, cycloalkyloxycarbonyl having 3 to 7 carbon atoms in the cycloalkyl moiety, or amino or aminocarbonyl. each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable amino substituents in each case being:

in each case straight-chain or branched alkyl having 1 to 4 carbon atoms, halogenoalkyl having 1 to 4 carbon atoms and 1 to 9 halogen atoms, alkoxyalkyl or alkylcarbonyl, each of which has 1 to 4 carbon atoms in the individual alkyl moieties, or arylcarbonyl, arylsulphonyl arylaminocarbonyl, or arylmethylsulphonyl, each of which has 6 or 10 carbon atoms in the aryl moiety and each of which is optionally monosubstituted to pentasubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>;

25

5

10

15

20

X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> furthermore represent aryl, arylthio, arylsulphinyl, arylsulphonyl, arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylthiomethylsulphonyl or arylazo, each of which has 6 or 10 carbon atoms in the aryl moiety, such as phenyl or naphthyl, and each of which is optionally monosubstituted to pentasubstituted in the aryl moiety by identical or different substituents, suitable aryl substituents in each case being those mentioned in the case of R<sup>1</sup>.

4. New substituted hetero-fused imidazoles of the general formula (I) according to Claim 1, characterized in that

represents hydrogen or a straight-chain or branched radical from the series consisting of alkyl and alkoxy, each of which has 1 to 4 carbon atoms and each of which is unsubstituted or substituted, or represents phenyl which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents being:

fluorine, chlorine, bromine, cyano, nitro, in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of which has 1 to 3 carbon atoms, in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl or halogenoalkylsulphonyl, each of which has 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, in each case straight-chain or branched alkoxyalkyl, alkoxyalkoxy, alkanoyl, alkoxycarbonyl or alkoximinoalkyl, each of which has 1 to 3 carbon atoms in the individual alkyl moieties, divalent dioxyalkylene having 1 to 3 carbon atoms which is optionally monosubstituted to tetrasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 3 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, or phenyl which is optionally monosubstituted to trisubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 3 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, halogen in each case representing fluorine, chlorine or bromine,

R<sup>2</sup> represents hydroxyl, cyano or a straight-chain or branched radical from the series consisting of alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy and dialkoxyphosphonyl, each of which has up to 4 carbon atoms in the

30

5

10

15

20

## 2158996

individual alkyl, alkenyl or alkinyl moieties and each of which is optionally monosubstituted to trisubstituted by identical or different substituents from the series consisting of fluorine, chlorine and bromine, or represents alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy or dialkoxyphosphoryl, each of which has up to 4 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties, and each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

10

5

straight-chain or branched alkoxy having 1 to 3 carbon atoms or phenyl which is optionally monosubstituted or disubstituted by identical or different substituents, suitable phenyl substituents being those mentioned in the case of R<sup>1</sup>,

15

R<sup>2</sup> furthermore represents amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

20

\_~

25

30

formyl, straight-chain or branched alkyl having 1 to 4 carbon atoms, straight-chain or branched alkenyl having 2 to 4 carbon atoms, straight-chain or branched alkylsulphonyl having 1 to 4 carbon atoms, carbamoyl, thiocarbamoyl or sulphamoyl, each of which is optionally monosubstituted or disubstituted by identical or different straight-chain or branched alkyl substituents having 1 to 4 carbon atoms, cycloalkyl, cycloalkylcarbonyl or cycloalkyloxycarbonyl, each of which has 3 to 6 carbon atoms in the cycloalkyl moiety, alkylcarbonyl, alkenylcarbonyl, alkoxycarbonyl, alkenyloxycarbonyl, alkylthio-carbonyl, alkoxy-thiocarbonyl or alkylthio-thiocarbonyl, each of which has 1 to 4 carbon atoms in the individual straight-chain or branched alkyl moieties, in each case divalent and cyclized alkanediylcarbonyl or alkanediyloxycarbonyl, each of which has 2 to 4 carbon atoms in the alkanediyl moiety, phenylalkyl, phenylalkylcarbonyl or phenylalkyloxycarbonyl, each of which has 1 to 4

carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or disubstituted in the phenyl moiety by identical or different substituents, or phenyl, phenylcarbonyl or phenyloxycarbonyl, each of which is optionally monosubstituted or disubstituted in the phenyl moiety by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of R<sup>1</sup>,

10

5

R<sup>2</sup> furthermore represents phenyl, phenylcarbonyl, phenylcarbonyloxy or phenylaminocarbonylaminocarbonyloxy, each of which is optionally monosubstituted to trisubstituted by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of R<sup>1</sup>,

15

R<sup>2</sup> furthermore represents heteroaryl, heteroarylcarbonyl, heteroaryloxy-carbonyl, heteroarylcarbonyloxy or heteroarylaminocarbonylaminocarbonyloxy, each of which have 2 to 9 carbon atoms and 1 to 3 identical or different hetero atoms (in particular nitrogen, oxygen and/or sulphur) in the heteroaryl moiety and each of which is optionally monosubstituted to trisubstituted by identical or different substituents, suitable heteroaryl substituents in each case being the phenyl substituents mentioned in the case of R<sup>1</sup>,

20

represents cyano, fluorine, chlorine, bromine, or a straight-chain or branched radical from the series consisting of alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl and alkylcarbonyloxy, each of which has up to 4 carbon atoms in the individual alkyl, alkenyl or alkinyl moieties and each of which is optionally monosubstituted to trisubstituted by identical or different substituents from the series consisting of fluorine, chlorine and bromine, or represents alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy or dialkoxyphosphoryl, each of which has up to 4 carbon atoms in the individual alkyl, alkenyl or alkinyl

30

moieties and each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

straight-chain or branched alkoxy having 1 to 3 carbon atoms or phenyl which is optionally monosubstituted or disubstituted by identical or different substituents, suitable phenyl substituents being those mentioned in the case of R<sup>1</sup>,

R³ furthermore represents amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable substituents in each case being:

formyl, straight-chain or branched alkyl having 1 to 4 carbon atoms, straight-chain or branched alkenyl having 2 to 4 carbon atoms, straightchain or branched alkylsulphonyl having 1 to 4 carbon atoms, in each case optionally monosubstituted or disubstituted (identically or differently by straight-chain or branched alkyl having 1 to 4 carbon atoms) carbamovl, thiocarbamoyl or sulphamoyl, cycloalkyl, cycloalkylcarbonyl, or cycloalkyloxycarbonyl, each of which has 3 to 6 carbon atoms in the cycloalkyl moiety, alkylcarbonyl, alkenylcarbonyl, alkoxycarbonyl, alkenyloxycarbonyl, alkylthio-carbonyl, alkoxy-thiocarbonyl or alkylthiothiocarbonyl, each of which has 1 to 4 carbon atoms in the individual straight-chain or branched alkyl moieties, in each case divalent and cyclized alkanediylcarbonyl or alkanediyloxycarbonyl, each of which has 2 to 4 carbon atoms in the alkanediyl moiety, phenylalkyl, phenylalkylcarbonyl or phenylalkyloxycarbonyl, each of which has 1 to 4 carbon atoms in the straight-chain or branched alkyl moiety and each of which is optionally monosubstituted or disubstituted in the phenyl moiety by identical or different substituents, or phenyl, phenylcarbonyl or phenyloxycarbonyl, each of which is optionally monosubstituted or disubstituted in the phenyl moiety by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case

30

5

10

15

20

of R1,

- R<sup>3</sup> furthermore represents phenyl which is optionally monosubstituted to trisubstituted by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of R<sup>1</sup>,
- A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> and A<sup>4</sup> in each case represent N(nitrogen), N-CHR<sup>1</sup>R<sup>2</sup> or CX, the hetero-fused ring having at least one, but not more than two, nitrogen atoms simultaneously and all positional isomers being possible, so that
  - CX1, CX2, CX3 exist in the case of one nitrogen atom and
  - CX<sup>1</sup> and CX<sup>2</sup> exist in the case of two nitrogen atoms, and, when either A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> or A<sup>4</sup> represent N-CHR<sup>1</sup>R<sup>2</sup>, the imidazole ring exists only in monosubstituted form, and
    - X1, X2 and X3 independently of one another in each case represent hydrogen, chlorine, bromine, cyano, nitro, in each case straight-chain or branched alkyl, alkoxy, alkylthio, alkylsulphinyl or alkylsulphonyl, each of which has 1 to 4 carbon atoms, cycloalkyl having 3, 5 or 6 carbon atoms, in each case straight-chain or branched halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl, halogenoalkylsulphonyl, each of which has 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, or represent divalent dioxyalkylene having 1 to 3 carbon atoms which is optionally monosubstituted to tetrasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 3 carbon atoms and straight-chain or branched halogenoalkyl having 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, furthermore represent hydroxycarbonyl, in each case straight-chain or branched alkylcarbonyl or alkoxycarbonyl, each of which has 1 to 3 carbon atoms in the alkyl moiety, cycloalkyloxycarbonyl having 3, 5 or 6 carbon atoms in the cycloalkyl moiety, or amino or aminocarbonyl, each of which is optionally monosubstituted or

25

10

15

phenylsulphonyl,

disubstituted by identical or different substituents, suitable amino substituents in each case being:

in each case straight-chain or branched alkyl having 1 to 3 carbon atoms, halogenoalkyl having 1 to 3 carbon atoms and 1 to 7 halogen atoms,

5

alkoxyalkyl or alkylcarbonyl, each of which has 1 to 3 carbon atoms in the individual alkyl moieties. phenylcarbonyl. or phenylaminocarbonyl or phenylmethylsulphonyl, each of which is optionally monosubstituted to trisubstituted in the phenyl moiety by

10

identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of R1;

X1, X2 and X3 furthermore represent phenyl, phenyloxy, phenylthio, phenylsulphinyl, phenylsulphonyl, phenylsulphonyloxy, phenylcarbonyl, phenyloxycarbonyl, phenylthiomethylsulphonyl or phenylazo, each of which is optionally monosubstituted to trisubstituted in the phenyl moiety by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of R<sup>1</sup>, and

15

20

25

30

where at least one of the substituents X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> represents in each case straight-chain halogenoalkyl, halogenoalkoxy, branched halogenoalkylthio, halogenoalkylsulphinyl, halogenoalkylsulphonyl, each of which has 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, or represents straight-chain or branched alkylsulphonyl having 1 to 3 carbon atoms, or represents divalent dioxyalkylene having 1 to 3 carbon atoms which is optionally monosubstituted to tetrasubstituted by identical or different substituents from the series consisting of halogen, straight-chain or branched alkyl having 1 to 3 carbon atoms and/or straight-chain or branched halogenoalkyl having 1 to 3 carbon atoms and 1 to 7 identical or different halogen atoms, furthermore represents hydroxycarbonyl, in each case straight-chain or branched alkylcarbonyl or alkoxycarbonyl, each of which has 1 to 3 carbon atoms in the alkyl moiety, cycloalkyloxycarbonyl having 3, 5 or 6 carbon atoms in the cycloalkyl moiety, or amino or aminocarbonyl, each of which is optionally monosubstituted or disubstituted by identical or different substituents, suitable amino substituents in each case being:

5

in each case straight-chain or branched alkyl having 1 to 3 carbon atoms, halogenoalkyl having 1 to 3 carbon atoms and 1 to 7 halogen atoms, alkoxyalkyl or alkylcarbonyl, each of which has 1 to 3 carbon atoms in the individual alkyl moieties, or phenylcarbonyl, phenylsulphonyl, phenylaminocarbonyl or phenylmethylsulphonyl, each of which is optionally monosubstituted to trisubstituted by identical or different substituents in the phenyl moiety, suitable phenyl substituents in each case being those mentioned in the case of R<sup>1</sup>;

10

15

- X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> furthermore represent phenyl, phenylthio, phenylsulphinyl, phenylsulphonyl, phenylsulphonyloxy, phenylcarbonyl, phenyloxycarbonyl, phenylthiomethylsulphonyl or phenylazo, each of which is optionally monosubstituted to trisubstituted in the phenyl moiety by identical or different substituents, suitable phenyl substituents in each case being those mentioned in the case of R<sup>1</sup>.
- 5. Process for the preparation of new substituted fused imidazoles of the general formula (I)

$$\begin{array}{c|c}
A & & & \\
\downarrow_{2} & & & \\
A & & & \\
A & & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\
& & & \\$$

20

in which

R<sup>1</sup> represents hydrogen or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl,

alkoxy and aryl,

R<sup>2</sup> represents hydroxyl, cyano or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl, alkenyl, alkinyl, alkoxy, alkenyloxy, alkinyloxy, alkylthio, amino, aminocarbonyl, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy, dialkoxyphosphonyl, (hetero)aryl, (hetero)aryl-carbonyl, (hetero)aryloxycarbonyl, (hetero)arylcarbonyloxy and (hetero)arylaminocarbonyl-aminocarbonyloxy,

R³ represents cyano, halogen or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl, alkenyl, alkinyl, alkylcarbonyl, alkoxycarbonyl, alkylcarbonyloxy, alkenyloxy, alkoxy, alkinyloxy, amino, aminocarbonyl and aryl,

A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> and A<sup>4</sup> in each case represent N(nitrogen), N-CHR<sup>1</sup>R<sup>2</sup> or CX, the hetero-fused ring having at least one, but not more than two, nitrogen atoms simultaneously and all positional isomers being possible, so that

CX1, CX2, CX3 exist in the case of one nitrogen atom and

CX<sup>1</sup> and CX<sup>2</sup> exist in the case of two nitrogen atoms, and, when either A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup> or A<sup>4</sup> represent N-CHR<sup>1</sup>R<sup>2</sup>, the imidazole ring exists only in monosubstituted form.

where,

X<sup>1</sup>, X<sup>2</sup> and X<sup>3</sup> in each case independently of one another represent hydrogen, halogen, cyano, nitro or a straight-chain or branched, in each case optionally unsubstituted or substituted, radical from the series consisting of alkyl, alkoxy, alkylthio, alkylsulphinyl, alkylsulphonyl and cycloalkyl, or represents hydroxycarbonyl, alkylcarbonyl, alkoxycarbonyl, cycloalkyloxycarbonyl, or represents in each case optionally substituted

25

5

10

amino or aminocarbonyl, or represents in each case optionally substituted aryl, aryloxy, arylthio, arylsulphinyl, arylsulphonyl, arylsulphonyl, arylcarbonyl, aryloxycarbonyl, arylazo or arylthiomethylsulphonyl, but where at least one of the substituents  $X^i$ ,  $X^2$  or  $X^3$  represents halogenoalkyl, halogenoalkoxy, halogenoalkylthio, halogenoalkylsulphinyl, halogenoalkylsulphonyl, alkylsulphonyl, or represents optionally substituted fused dioxyalkylene, or represents hydroxycarbonyl, alkylcarbonyl, alkoxycarbonyl, cycloalkyloxycarbonyl, or represents in each case optionally substituted amino or aminocarbonyl, or represents in each case optionally substituted aryl, arylthio, arylsulphinyl, arylsulphonyl, arylsulphonyloxy, arylcarbonyl, aryloxycarbonyl, arylazo or arylthiomethylsulphonyl,

characterized in that

1H-substituted hetero-fused imidazoles of the formula (II)

$$\begin{array}{c}
A^{3} \\
\downarrow_{2} \\
A \\
A
\end{array}$$

$$\begin{array}{c}
N \\
H$$

$$\begin{array}{c}
N \\
H
\end{array}$$

in which

5

10

A<sup>1</sup>, A<sup>2</sup>, A<sup>3</sup>, A<sup>4</sup> and R<sup>3</sup> have the abovementioned meanings

are reacted with compounds of the formula (III)

in which

M represents a suitable leaving group and

R1 and R2 have the abovementioned meanings,

if appropriate in the presence of a diluent and if appropriate in the presence of a reaction auxiliary.

- Herbicidal compositions, characterized in that they comprise at least one substituted hetero-fused imidazole of the formula (I) according to Claims 1 to
   5.
- Method of combating undesirable plants, characterized in that substituted hetero-fused imidazoles of the general formula (I) according to Claims 1 to 5 are allowed to act on plants and/or their environment.
  - 8. Use of substituted, hetero-fused imidazoles of the general formula (I) according to Claims 1 to 5 for combating undesirable plants.
- Process for the preparation of herbicidal and insecticidal compositions, characterized in that substituted hetero-fused imidazoles of the general formula
   (I) according to Claims 1 to 5 are mixed with extenders and/or surface-active substances.
  - Use of substituted hetero-fused imidazoles of the general formula (I) according to Claims 1 to 5 for combating animal pests.

Fetherstonhaugh & Co., Ottawa, Canada Patent Agents